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Your Ref:
Our Ref: AFB/HJ/62619A
Our Case No. P10392EP-W

16th May 2005

Dear Sirs,

Re: New European Patent Application No. 03768952.8
(ex PCT/US2003/036483)
- DOW GLOBAL TECHNOLOGIES INC.

Please mark the file of this application such that all correspondence is directed to me at the address shown below. John Raynor is also an authorised representative, but not the one to whom correspondence in respect of the application should be directed.

Yours faithfully,

Anthony F. Burford
Authorised European Patent Attorney
(e-mail: afburford@beckgreener.com)

US 03/36483 (A41) printer

EP1572757 - OLEFIN POLYMERIZATION CATALYST COMPOSITION COMPRISING GROUP 13 AMIDE DERIVATIVES - DOW GLOBAL TECHNOLOGIES INC.

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01/07/2004	International publication pamphlet	Search/Examination	30
01/07/2004	Copy of the International Search report	Search/Examination	3
26/07/2004	Priority document (electronically filed)	Search/Examination	32
03/05/2005	Request to enter regional phase (EPCT)	Search/Examination	10
06/05/2005	Request to enter regional phase (EPCT)	Search/Examination	6
06/05/2005	Matter concerning the application	Search/Examination	1
18/05/2005	Copy of the International preliminary examination report	Search/Examination	5
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Total Number of Pages: 88

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PROVISIONAL APPLICATION COVER SHEET

This is a request for filing a PROVISIONAL APPLICATION under 35 CFR 1.53 (c).

Docket Number	62619	Type a plus sign (+) inside this box

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** ☐ *Additional inventors are being named on separately numbered sheets attached hereto***

TITLE OF THE INVENTION (280 characters max)

OLEFIN POLYMERIZATION CATALYST COMPOSITION COMPRISING GROUP 13 AMIDE DERIVATIVES

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Respectfully submitted,

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OLEFIN POLYMERIZATION CATALYST COMPOSITION COMPRISING GROUP 13 AMIDE DERIVATIVES

Background of the Invention

This invention relates to compositions of matter which are useful as addition polymerization catalysts, to a method for preparing these catalyst compositions and to a method of using these catalyst compositions. More particularly, this invention relates to improved olefin polymerization catalyst compositions comprising a Group 4 metal complex, an activator therefor, and a Group 13 aromatic amide tertiary component. The invention also relates to an improved method for polymerizing addition polymerizable monomers using these catalyst compositions.

Constrained geometry metal complexes, their preparation, methods of activation, active catalysts formed therefrom including cationic catalysts and methods of use are disclosed in EP-A-416,815; EP-A-514,828; EP-A-520,732; US-A-5.064,802; US-A-5,374,696; USP 5,470,993; US-A-5,055,438, US-A-5,057,475, US-A-5,096,867, US-A-5,064,802, US-A-5,132,380, and US-A-5,453,410.

Although previously known active catalysts, especially the cationic catalysts disclosed in the foregoing applications and publications, have excellent activity they are extremely sensitive to catalyst poisons, such as polar impurities, that may be contained in a polymerization mixture. Because of this fact, catalyst efficiencies and lifetimes have been limited and molecular weights of the resulting polymers have been reduced. In addition, it has now been observed that certain tertiary substances that are added to the reaction mixture to improve catalyst efficiency disadvantageously result in the formation of increased levels of a polymer fraction having high crystallinity. Such high crystallinity fractions (HCF) may result in fouling of the reactor and are desirably eliminated or at least reduced in order to increase production efficiency and product uniformity.

It is previously known in the art to utilize adjuvants such as trialkylboron compounds, trialkylaluminum compounds, dialkylaluminum alkoxides and dialkylaluminum N,N-di(hydrocarbyl)amides to remove catalyst poisons from various olefin polymerization catalysts. Examples of such compositions are contained in USA-6,074,977, USA-6,017,842, USA-5,206,199, USA-5,962,599, USA-6,268,063 and USA-6,353063. Disadvantageously however, such adjuvants have proven to be less effective in combating the inhibition of catalytically activated constrained geometry catalysts, and have not demonstrated reduced HCF formation or reduced reactor fouling.

The present investigations have led to certain improved catalyst compositions that are highly active as addition polymerization catalysts, desirably having improved resistance to catalyst poisons with reduced production of high crystalline fraction polymers.

Summary of the Invention

According to the present invention there is now provided a catalyst composition comprising:

a) a transition metal complex capable of being activated for polymerization of addition polymerizable monomers;

b) an activator compound able to render the transition metal complex catalytically active for polymerization of addition polymerizable monomers; and

c) a Group 13 metal compound corresponding to the formula:

$$R^aM(N(R^b)_2)_2 \text{ (1)} \quad \text{or} \quad \begin{matrix} R^d\text{—}N(R^b) \\ | \quad\quad | \\ R^c{}_2M \quad MR^c{}_2 \end{matrix} \text{ (2)};$$

wherein,

M, independently each occurrence is a group 13 metal, preferably aluminum;

R^a is a hydrocarbyl, halocarbyl, halohydrocarbyl, tri(hydrocarbyl)silyl, or tri(hydrocarbyl)silyl- substituted hydrocarbyl radical of from 1 to 20 carbon, silicon or mixtures of carbon and silicon atoms, preferably C_{1-6} alkyl;

R^b independently each occurrence is a C_{1-30} hydrocarbyl group, preferably alkyl or aryl, most preferably C_{6-20} aryl;

R^c independently each occurrence is selected from hydrogen, R^a, $-NR^b{}_2$, or a halo- or di(C_{1-10} hydrocarbyl)amino- substituted hydrocarbyl group, and optionally one or more of R^c groups may be shared by both metal centers, M, in the form of a μ-bridged structure, preferably R^c is a hydrocarbyl group or $-NR^b{}_2$, wherein R^b is C_{6-20} aryl; and

R^d, is a divalent, anionic ligand group of up to 30 atoms, not counting hydrogen, preferably a hydrocarbadiyl group, or a halo- or di(C_{1-10} hydrocarbyl)amino- substituted hydrocarbadiyl group, most preferably R^d is C_{6-20} arylene.

Further according to the present invention there is provided a process for polymerization of addition polymerizable monomers or mixtures thereof comprising contacting said monomer or mixture of monomers with a catalyst system comprising the above catalyst composition under addition polymerization conditions. Preferred addition polymerizable monomers include $C_{2-20,000}$ α-olefins. Polymers prepared by the foregoing

invented process are usefully employed for molding, film, sheet, extrusion foaming and other applications.

As a final embodiment of the invention, there is provided the foregoing group 13 compounds (2), which are novel compounds that are usefully employed as tertiary polymerization components in the foregoing process.

Use of the present catalyst compositions and processes result in the highly efficient production of high molecular weight olefin polymers over a wide range of polymerization conditions, with reduced incidence of high crystalline fraction polymer formation. They are especially useful for the formation of copolymers of ethylene and styrene (ES polymers) and ethylene/styrene/diene (ESDM polymers) wherein the diene is ethylidenenorbornene, 1,4-hexadiene or similar nonconjugated diene.

The catalyst compositions of this invention may also be supported on a support material and used in olefin polymerization processes in a slurry or in the gas phase. The catalyst may be prepolymerized with one or more olefin monomers *in situ* in a polymerization reactor or in a separate process with intermediate recovery of the prepolymerized catalyst prior to the primary polymerization process.

Brief Description of the Drawings

Figure 1 is a computer rendering (ORTEP) of the metal complex of Example 1 determined by single crystal X-ray diffraction analysis.

Detailed Description of the Invention

All reference to the Periodic Table of the Elements herein shall refer to the Periodic Table of the Elements, published and copyrighted by CRC Press, Inc., 2001. Also, any reference to a Group or Groups shall be to the Group or Groups as reflected in this Periodic Table of the Elements using the IUPAC system for numbering groups. For purposes of United States patent practice, the contents of any patent, patent application or publication referenced herein is hereby incorporated by reference in its entirety, especially with respect to the disclosure of analytical or synthetic techniques and general knowledge in the art.

The term "comprising" and derivatives thereof is not intended to exclude the presence of any additional component, step or procedure, whether or not the same is disclosed herein. In order to avoid any doubt, all compositions claimed herein through use of the term "comprising" may include any additional additive, adjuvant, or compound whether polymeric or otherwise, unless stated to the contrary. In contrast, the term, "consisting essentially of" excludes from the scope of any succeeding recitation any other component, step or procedure,

excepting those that are not essential to operability. The term "consisting of" excludes any component, step or procedure not specifically delineated or listed. The term "or", unless stated otherwise, refers to the listed members individually as well as in any combination.

The term "polymer", as used herein, includes both homopolymers, that is, polymers prepared from a single reactive compound, and copolymers, that is, polymers prepared by reaction of at least two polymer forming reactive, monomeric compounds. The term "crystalline" refers to a polymer that exhibits an X-ray diffraction pattern at 25 °C and possesses a first order transition or crystalline melting point (Tm). The term may be used interchangeably with the term "semicrystalline".

The foregoing components a), b) and c) of the catalyst composition are present in relative amounts to produce polymeric materials in a highly efficient polymerization process. Highly desirably, the molar ratio of a):b) is from 1:0.1 to 1:1000, more preferably from 1:0.5 to 1:100. The molar ratio of a):c) is desirably from 1:0.1 to 1:10, more preferably from 1:0.5 to 1:1. The respective components may be combined in any order or incorporated into other components of the reaction mixture such as the solvent or monomer(s) prior to contacting with the remaining components of the catalyst composition. Highly desirably, component c) is added to the solvent or monomer(s) employed in the polymerization prior to contacting with the remaining catalyst components a) and c).

The catalyst, component a), is desirably a metal complex corresponding to the formula:

$$L_tMX_pX'_q \quad \text{(II)}$$

wherein: M is a metal of Group 4 of the Periodic Table of the Elements having an oxidation state of +2, +3 or +4, bound in an η^5 bonding mode to one or more L groups;

L independently each occurrence is a cyclopentadienyl-, indenyl-, tetrahydroindenyl-, fluorenyl-, tetrahydrofluorenyl-, or octahydrofluorenyl- group optionally substituted with from 1 to 8 substituents independently selected from the group consisting of hydrocarbyl, halo, halohydrocarbyl, aminohydrocarbyl, hydrocarbyloxy, dihydrocarbylamino, dihydrocarbylphosphino, silyl, aminosilyl, hydrocarbyloxysilyl, and halosilyl groups containing up to 20 non-hydrogen atoms, or further optionally two such L groups may be joined together by a divalent substituent selected from hydrocarbadiyl, halohydrocarbadiyl, hydrocarbyleneoxy, hydrocarbyleneamino, siladiyl, halosiladiyl, and divalent aminosilane, groups containing up to 20 non-hydrogen atoms;

X independently each occurrence is a monovalent or polyvalent anionic ligand group having one or more shared of donative bonds to M, and optionally one or more shared or donative bonds to one or more L groups, said X containing up to 60 nonhydrogen atoms;

X' independently each occurrence is a neutral Lewis base ligating compound, having up to 20 atoms;

t, p, and q are 0, 1 or 2.

The compositions of the present invention are believed to exist in the form of a mixture of one or more cationic, zwitterionic or other catalytically active species derived from the foregoing metal complex a) in combination with the activator compound, b), or alternatively, a mixture of the metal complex or a cationic, zwitterionic or other catalytically active derivative thereof with a derivative formed by interaction of compound c) with the cocatalyst or with the activated catalyst. Fully cationic or partially charge separated metal complexes, that is, zwitterionic metal complexes, have been previously disclosed in US-A-5,470,993 and 5,486,632. Derivatives of the Group 13 compound and cocatalyst may arise, for example, by ligand exchange. In particular, where the cocatalyst is a strong Lewis acid, such as tris(fluorophenyl)borane, some quantity of fluorophenyl substituents may exchange with the ligand groups of the Group 13 compound to form fluorophenyl substituted derivatives thereof.

The cationic complexes are believed to correspond to the formula: $L_tM^+X_{p-1}A^-$ (III) wherein:

M is a Group 4 metal in the +4 or +3 formal oxidation state;

L, X, t and p are as previously defined; and

A^- is a noncoordinating, compatible anion derived from the activating cocatalyst.

The zwitterionic complexes in particular result from activation of a Group 4 metal diene complex that is in the form of a metallocyclopentene, wherein the metal is in the +4 formal oxidation state, (that is X is 2-butene-1,4-diyl, or a hydrocarbyl substituted derivative thereof, having both valencies bonded to M) by the use of a Lewis acid activating cocatalyst, especially tris(perfluoroaryl)boranes. These zwitterionic complexes are believed to correspond to the formula: $L_tM^+X_{p-1}X^{**}\text{-}A^-$ (IV)

wherein:

M is a Group 4 metal in the +4 formal oxidation state;

L, X, t and p are as previously defined;

X^{**} is the divalent remnant of the conjugated diene, X', formed by ring opening at one of the carbon to metal bonds of a metallocyclopentene; and

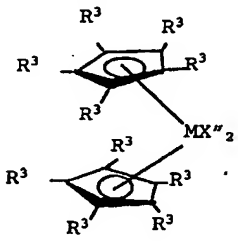
A^- is a noncoordinating, compatible anion derived from the activating cocatalyst.

As used herein, the recitation "noncoordinating" means an anion which either does not coordinate to component a) or which is only weakly coordinated therewith remaining

sufficiently labile to be displaced by a neutral Lewis base, including an α-olefin. A non-coordinating anion specifically refers to an anion which when functioning as a charge balancing anion in the catalyst system of this invention, does not transfer a fragment thereof to said cation thereby forming a neutral four coordinate metal complex and a neutral byproduct. "Compatible anions" are anions which are not degraded to neutrality when the initially formed complex decomposes and are noninterfering with desired subsequent polymerizations.

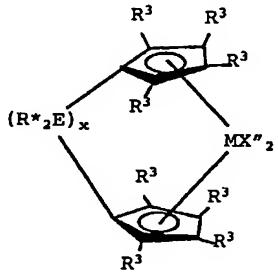
Preferred X' groups are phosphines, especially trimethylphosphine, triethylphosphine, triphenylphosphine and bis(1,2-dimethylphosphino)ethane; $P(OR)_3$, wherein R is as previously defined; ethers, especially tetrahydrofuran; amines, especially pyridine, bipyridine, tetramethylethylenediamine (TMEDA), and triethylamine; olefins; and conjugated dienes having from 4 to 40 carbon atoms. Complexes including conjugated diene X' groups include those wherein the metal is in the +2 formal oxidation state.

Examples of coordination complexes a) used according to the present invention include the foregoing species:



(V)

or



(VI)

wherein:

M is titanium, zirconium or hafnium, preferably zirconium or hafnium, in the +2 or +4 formal oxidation state;

R^3 in each occurrence independently is selected from the group consisting of hydrogen, hydrocarbyl, silyl, germyl, cyano, halo and combinations thereof, said R^3 having up to 20 non-hydrogen atoms, or adjacent R^3 groups together form a divalent derivative (i.e., a hydrocarbadiyl, siladiyl or germadiyl group) thereby forming a fused ring system,

X" independently each occurrence is an anionic ligand group of up to 40 non-hydrogen atoms, or two X" groups together form a divalent anionic ligand group of up to 40 non-hydrogen atoms or together are a conjugated diene having from 4 to 30 non-hydrogen atoms forming a π-complex with M, whereupon M is in the +2 formal oxidation state,

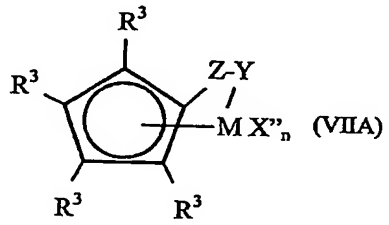
R* independently each occurrence is C_{1-4} alkyl or phenyl,

E independently each occurrence is carbon or silicon, and

x is an integer from 1 to 8.

Additional examples of metal complexes a) include those corresponding to the formula: $LMX_pX'_q$ (VII)

wherein L, M, X, X', p and q are as previously defined. A preferred metal complex belongs to the foregoing class (VII) and corresponds to the formula:



(VIIA)

wherein:

M is titanium, zirconium or hafnium in the +2, +3 or +4 formal oxidation state;

R^3 in each occurrence independently is selected from the group consisting of hydrogen, hydrocarbyl, silyl, germyl, cyano, halo and combinations thereof, said R^3 having up to 20 non-hydrogen atoms, or adjacent R^3 groups together form a divalent derivative (i.e., a hydrocarbadiyl, siladiyl or germadiyl group) thereby forming a fused ring system,

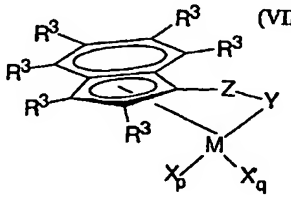
each X" is a halo, hydrocarbyl, hydrocarbyloxy, hydrocarbylamino, or silyl group, said group having up to 20 non-hydrogen atoms, or two X" groups together form a neutral C_{5-30} conjugated diene or a divalent derivative thereof;

Y is -O-, -S-, -NR*-, -PR*-;

Z is SiR^*_2, CR^*_2, $SiR^*_2SiR^*_2$, $CR^*_2CR^*_2$, $CR^*=CR^*$, $CR^*_2SiR^*_2$, or GeR^*_2, wherein R* is as previously defined, and

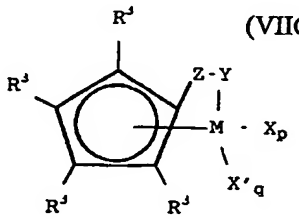
n is an integer from 1 to 3.

Most preferred coordination complexes a) used according to the present invention are complexes corresponding to the formula:



(VIIB)

or



(VIIC)

wherein:

R^3 independently each occurrence is a group selected from hydrogen, hydrocarbyl, halohydrocarbyl, silyl, germyl and mixtures thereof, said group containing up to 20 nonhydrogen atoms;

M is titanium, zirconium or hafnium;

Z, Y, X and X' are as previously defined;

p is 0, 1 or 2; and

q is zero or one;

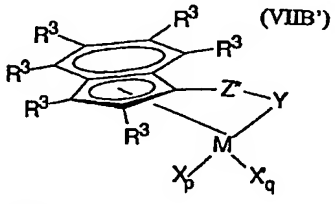
with the proviso that:

when p is 2, q is zero, M is in the +4 formal oxidation state, and X is an anionic ligand selected from the group consisting of halide, hydrocarbyl, hydrocarbyloxy, di(hydrocarbyl)amido, di(hydrocarbyl)phosphido, hydrocarbylsulfido, and silyl groups, as well as halo-, di(hydrocarbyl)amino-, hydrocarbyloxy- and di(hydrocarbyl)-phosphino-substituted derivatives thereof, said X group having up to 20 nonhydrogen atoms,

when p is 1, q is zero, M is in the +3 formal oxidation state, and X is a stabilizing anionic ligand group selected from the group consisting of allyl, 2-(N,N-dimethylaminomethyl)phenyl, and 2-(N,N-dimethyl)-aminobenzyl, or M is in the +4 formal oxidation state, and X is a divalent derivative of a conjugated diene, M and X together forming a metallocyclopentene group, and

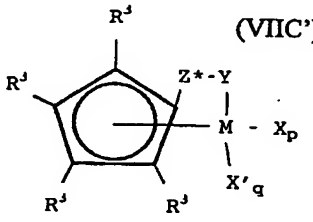
when p is 0, q is 1, M is in the +2 formal oxidation state, and X' is a neutral, conjugated or nonconjugated diene, optionally substituted with one or more hydrocarbyl groups, said X' having up to 40 carbon atoms and forming a π-complex with M.

More preferred coordination complexes a) used according to the present invention are complexes corresponding to the formula:



(VIIB')

or



(VIIC')

wherein:

R^3 independently each occurrence is hydrogen or C_{1-6} alkyl;

M is titanium;

Y is -O-, -S-, -NR*-, -PR*-;

Z* is SiR^*_2, CR^*_2, $SiR^*_2SiR^*_2$, $CR^*_2CR^*_2$, $CR^*=CR^*$, $CR^*_2SiR^*_2$, or GeR^*_2;

R* each occurrence is independently hydrogen, or a member selected from hydrocarbyl, hydrocarbyloxy, silyl, halogenated alkyl, halogenated aryl, and combinations thereof, said R* having up to 20 non-hydrogen atoms, and optionally, two R* groups from Z (when R* is not hydrogen), or an R* group from Z and an R* group from Y form a ring system;

p is 0, 1 or 2;

q is zero or one;

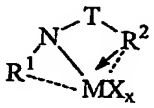
with the proviso that:

when p is 2, q is zero, M is in the +4 formal oxidation state, and X is independently each occurrence methyl or benzyl,

when p is 1, q is zero, M is in the +3 formal oxidation state, and X is 2-(N,N-dimethyl)aminobenzyl; or M is in the +4 formal oxidation state and X is 2-butene-1,4-diyl, and

when p is 0, q is 1, M is in the +2 formal oxidation state, and X' is 1,4-diphenyl-1,3-butadiene or 1,3-pentadiene. The latter diene is illustrative of unsymetrical diene groups that result in production of metal complexes that are actually mixtures of the respective geometrical isomers.

Additional examples of suitable metal complexes for use in the present invention include Group 4 metal derivatives, especially hafnium derivatives of hydrocarbylamine substituted heteroaryl compounds of the formula $R^1HN\text{-}T\text{-}R^2$ (VII), said complexes corresponding to the formula:



(VIIA)

wherein

R^1 is selected from alkyl, cycloalkyl, heteroalkyl, cycloheteroalkyl, aryl, and inertly substituted derivatives thereof containing from 1 to 30 atoms not counting hydrogen;

T is a divalent bridging group of from 1 to 20 atoms other than hydrogen, preferably a mono- or di- C_{1-20} hydrocarbyl substituted methylene or silane group, and

R^2 is a C_{6-20} heteroaryl group containing Lewis base functionality, especially a pyridin-2-yl- or substituted pyridin-2-yl group,

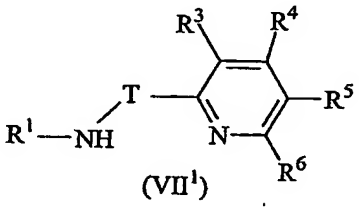
and in the metal complex, M is the Group 4 metal, preferably hafnium,

X is an anionic, neutral or dianionic ligand group,

x is a number from 0 to 5 indicating the number of such X groups, and

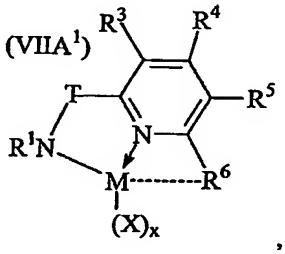
bonds, optional bonds and electron donative interactions are represented by lines, dotted lines and arrows respectively.

Preferred complexes are those wherein ligand formation results from hydrogen elimination from the amine group and optionally from the loss of one or more additional groups, especially from R^2. In addition, electron donation from the Lewis basic, heteroaryl functionality, preferably an electron pair, provides additional stability to the metal center. Preferred examples of the foregoing polyfunctional Lewis base compounds and the resulting metal complexes correspond to the formulas:



(VII^1)

and



$(VIIA^1)$

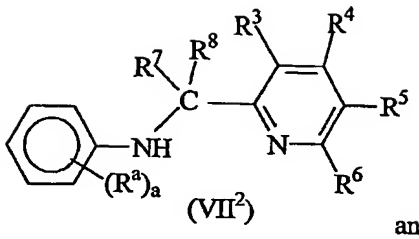
, wherein

M, X, x, R^1 and T are as previously defined,

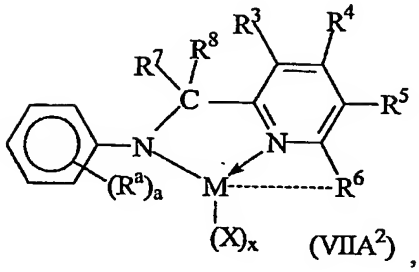
R^3, R^4, R^5 and R^6 are hydrogen, halo, or an alkyl, cycloalkyl, heteroalkyl, heterocycloalkyl, aryl, or silyl group of up to 20 atoms not counting hydrogen, or adjacent R^3, R^4, R^5 or R^6 groups may be joined together thereby forming fused ring derivatives, and

bonds, optional bonds and electron pair donative interactions are represented by lines, dotted lines and arrows respectively.

More preferred examples of the foregoing difunctional Lewis base compounds and metal complexes correspond to the formula:



(VII^2) and



(VIIA^2),

wherein

M, X, x, R^1 and T are as previously defined,

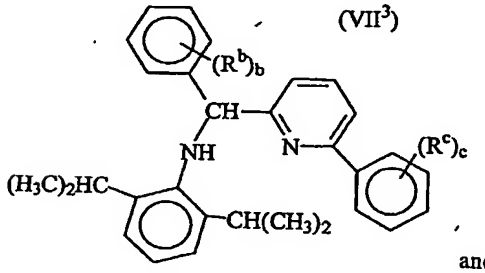
R^3, R^4, R^5 and R^6 are as previously defined, preferably R^3, R^4, and R^5 are hydrogen, or C_{1-4} alkyl, and R^6 is C_{6-20} aryl, most preferably naphthalenyl;

R^a independently each occurrence is C_{1-4} alkyl, and a is 1-5, most preferably R^a in two ortho- positions is isopropyl or t-butyl;

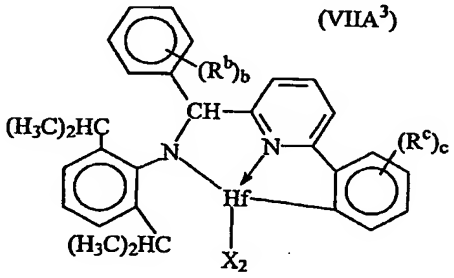
R^7 and R^8 independently each occurrence are hydrogen or a C_{1-20} alkyl or aryl group, most preferably one of R^7 and R^8 is hydrogen and the other is a C_{6-20} aryl group, especially a fused polycyclic aryl group, most preferably an anthracenyl group, and

bonds, optional bonds and electron pair donative interactions are represented by lines, dotted lines and arrows respectively.

Highly preferred polyfunctional Lewis base compounds and metal complexes for use herein correspond to the formula:



(VII^3) and



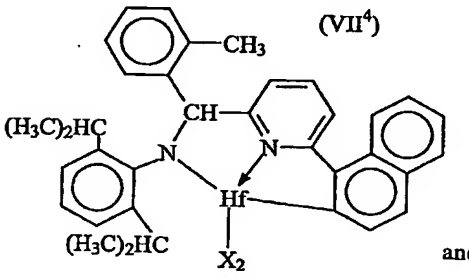
(VIIA^3),

wherein X each occurrence is halide, N,N-dimethylamido, or C_{1-4} alkyl, and preferably each occurrence X is methyl;

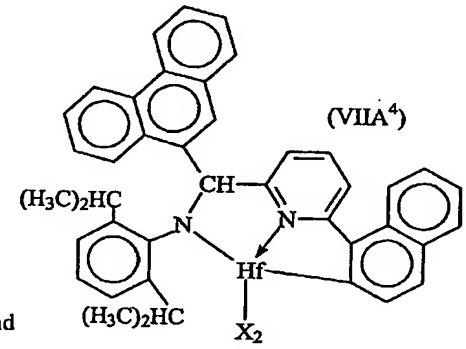
R^b independently each occurrence is C_{1-20} alkyl or aryl, or two adjacent R^b groups are joined together thereby forming a ring, and b is 1-5; and

R^c independently each occurrence is C_{1-20} alkyl or aryl, or two adjacent R^c groups are joined together thereby forming a ring, and c is 1-5.

Most highly preferred examples of metal complexes for use according to the present invention are complexes of the following formulas:



and



wherein X each occurrence is halide, N,N-dimethylamido, or C_{1-4} alkyl, and preferably each occurrence X is methyl.

Specific examples of metal complexes (catalysts) usefully employed as component a) according to the present invention include:

bis(cyclopentadienyl)zirconiumdimethyl,
bis(cyclopentadienyl)zirconium dibenzyl,
bis(cyclopentadienyl)zirconium methyl benzyl,
bis(cyclopentadienyl)zirconium methyl phenyl,
bis(cyclopentadienyl)zirconiumdiphenyl,
bis(cyclopentadienyl)titanium-allyl,
bis(cyclopentadienyl)zirconiummethylmethoxide,
bis(cyclopentadienyl)zirconiummethylchloride,
bis(pentamethylcyclopentadienyl)zirconiumdimethyl,
bis(pentamethylcyclopentadienyl)titaniumdimethyl,
bis(indenyl)zirconiumdimethyl,
indenylfluorenylzirconiumdimethyl,
bis(indenyl)zirconiummethyl(2-(dimethylamino)benzyl),
bis(indenyl)zirconiummethyltrimethylsilyl,
bis(tetrahydroindenyl)zirconiummethyltrimethylsilyl,
bis(pentamethylcyclopentadienyl)zirconiummethylbenzyl,
bis(pentamethylcyclopentadienyl)zirconiumdibenzyl,

bis(pentamethylcyclopentadienyl)zirconiummethylmethoxide,
bis(pentamethylcyclopentadienyl)zirconiummethylchloride,
bis(methylethylcyclopentadienyl)zirconiumdimethyl,
bis(butylcyclopentadienyl)zirconiumdibenzyl,
bis(t-butylcyclopentadienyl)zirconiumdimethyl,
bis(ethyltetramethylcyclopentadienyl)zirconiumdimethyl,
bis(methylpropylcyclopentadienyl)zirconiumdibenzyl,
bis(trimethylsilylcyclopentadienyl)zirconiumdibenzyl,
dimethylsilyl-bis(cyclopentadienyl)zirconiumdimethyl,
dimethylsilyl-bis(tetramethylcyclopentadienyl)titanium (III) allyl
dimethylsilyl-bis(t-butylcyclopentadienyl)zirconiumdibenzyl,
dimethylsilyl-bis(n-butylcyclopentadienyl)zirconium bis(trimethylsilyl),
(methylene-bis(tetramethylcyclopentadienyl)titanium(III) 2-(dimethylamino)benzyl,
(methylene-bis(n-butylcyclopentadienyl)titanium(III) 2-(dimethylamino)benzyl,
dimethylsilyl-bis(indenyl)zirconiumbenzylchloride,
dimethylsilyl-bis(2-methylindenyl)zirconiumdimethyl,
dimethylsilyl-bis(2-methyl-4-phenylindenyl)zirconiumdimethyl,
dimethylsilyl-bis(2-methylindenyl)zirconium-1,4-diphenyl-1,3-butadiene,
dimethylsilyl-bis(2-methyl-4-phenylindenyl)zirconium (II) 1,4-diphenyl-1,3-butadiene,
dimethylsilyl-bis(tetrahydroindenyl)zirconium(II) 1,4-diphenyl-1,3-butadiene,
di(isopropylamino)borandiylbis(2-methyl-4-phenylindenyl)zirconium dimethyl,
dimethylsilyl-bis(tetrahydrofluorenyl)zirconium bis(trimethylsilyl),
(isopropylidene)(cyclopentadienyl)(fluorenyl)zirconiumdibenzyl,
dimethylsilyl(tetramethylcyclopentadienyl)(fluorenyl)zirconium dimethyl,
cyclopentadienyltitaniumtrimethyl,
indenyltitaniumtrimethyl,
octahydrofluorenyltitaniumtrimethyl,
tetrahydroindenyltitaniumtrimethyl,
tetrahydrofluorenyltitaniumtrimethyl,
(tert-butylamido)(tetramethyl-η^5-cyclopentadienyl) dimethylsilanetitanium dibenzyl,
(tert-butylamido)(tetramethyl-η^5-cyclopentadienyl)dimethylsilanetitanium dimethyl,
(tert-butylamido)(tetramethyl-η^5-cyclopentadienyl)-1,2-ethanediyltitanium dimethyl,
(tert-butylamido)(tetramethyl-η^5-indenyl)dimethylsilanetitanium dimethyl,
(tert-butylamido)(tetramethyl-η^5-cyclopentadienyl)dimethylsilane titanium (III)

2-(dimethylamino)benzyl;

(tert-butylamido)(tetramethyl-η^5-cyclopentadienyl)dimethylsilanetitanium (III) allyl,

(tert-butylamido)(tetramethyl-η^5-cyclopentadienyl)dimethylsilanetitanium (III) 2,4-dimethylpentadienyl,

(tert-butylamido)(tetramethyl-η^5-cyclopentadienyl)dimethylsilanetitanium (II) 1,4-diphenyl-1,3-butadiene,

(tert-butylamido)(tetramethyl-η^5-cyclopentadienyl)dimethylsilanetitanium (II) 1,3-pentadiene,

(tert-butylamido)(2-methylindenyl)dimethylsilanetitanium (II) 1,4-diphenyl-1,3-butadiene,

(tert-butylamido)(2-methylindenyl)dimethylsilanetitanium (II) 2,4-hexadiene,

(tert-butylamido)(2-methylindenyl)dimethylsilanetitanium (IV) 2,3-dimethyl-1,3-butadiene,

(tert-butylamido)(2-methylindenyl)dimethylsilanetitanium (II) 1,3-pentadiene,

(tert-butylamido)(2-methylindenyl)dimethylsilanetitanium (IV) 1,3-butadiene,

(tert-butylamido)(2,3-dimethylindenyl)dimethylsilanetitanium (IV) 2,3-dimethyl-1,3-butadiene,

(tert-butylamido)(2,3-dimethylindenyl)dimethylsilanetitanium (IV) isoprene

(tert-butylamido)(2,3-dimethylindenyl)dimethylsilanetitanium (IV) dimethyl

(tert-butylamido)(2,3-dimethylindenyl)dimethylsilanetitanium (IV) dibenzyl

(tert-butylamido)(2,3-dimethylindenyl)dimethylsilanetitanium (IV) 1,3-butadiene,

(tert-butylamido)(2,3-dimethylindenyl)dimethylsilanetitanium (II) 1,3-pentadiene,

(tert-butylamido)(2,3-dimethylindenyl)dimethylsilanetitanium (II) 1,4-diphenyl-1,3-butadiene,

(tert-butylamido)(2-methyl-(*s*)-indacenyl)dimethylsilanetitanium (II) 1,3-pentadiene,

(tert-butylamido)(2-methyl-(*s*)-indacenyl)dimethylsilanetitanium (IV) dimethyl,

(tert-butylamido)(2-methyl-(*s*)-indacenyl)dimethylsilanetitanium (IV) dibenzyl,

(tert-butylamido)(2-methyl-4-phenylindenyl)dimethylsilanetitanium (II) 1,4-diphenyl-1,3-butadiene,

(tert-butylamido)(2-methyl-4-phenylindenyl)dimethylsilanetitanium (II) 1,3-pentadiene,

(tert-butylamido)(2-methyl-4-phenylindenyl)dimethylsilanetitanium (II) 2,4-hexadiene,

(cyclohexylamido)(tetramethyl-η^5-cyclopentadienyl)dimethylsilanetitanium (IV) dimethyl,

(tert-butylamido)(tetramethyl-η^5-cyclopentadienyl)dimethylsilanetitanium (IV) 2,3-dimethyl-1,3-butadiene,

(tert-butylamido)(tetramethyl-η^5-cyclopentadienyl)bis(4-dodecylphenyl)silanetitanium (IV) dimethyl,

(tert-butylamido)(tetramethyl-η^5-cyclopentadienyl)bis(4-dodecylphenyl)silanetitanium (II) 1,4-diphenyl-1,3-butadiene,

(tert-butylamido)(tetramethyl-η^5-cyclopentadienyl)bis(4-dodecylphenyl)silanetitanium (II) 1,3-pentadiene,

(tert-butylamido)(3-(N-pyrrolyl)indenyl)dimethylsilanetitanium (IV) 2,3-dimethyl-1,3-butadiene,

(tert-butylamido)(3-(N-pyrrolyl)indenyl)dimethylsilanetitanium (IV) isoprene

(tert-butylamido)(3-(N-pyrrolyl)indenyl)dimethylsilanetitanium (IV) dimethyl

(tert-butylamido)(3-(N-pyrrolyl)indenyl)dimethylsilanetitanium (IV) dibenzyl

(tert-butylamido)(3-(N-pyrrolyl)indenyl)dimethylsilanetitanium (IV) 1,3-butadiene,

(tert-butylamido)(3-(N-pyrrolyl)indenyl)dimethylsilanetitanium (II) 1,3-pentadiene,

(tert-butylamido)(3-(N-pyrrolyl)indenyl)dimethylsilanetitanium (II) 1,4-diphenyl-1,3-butadiene,

(tert-butylamido)(3-N-pyrrolidinylinden-1-yl)dimethylsilanetitanium (IV) dimethyl,

[N-(2,6-di(1-methylethyl)phenyl)amido)(o-tolyl)(α-naphthalen-2-diyl(6-pyridin-2-diyl)methane)]hafnium dimethyl,

[N-(2,6-di(1-methylethyl)phenyl)amido)(o-tolyl)(α-naphthalen-2-diyl(6-pyridin-2-diyl)methane)]hafnium di(N,N-dimethylamido),

[N-(2,6-di(1-methylethyl)phenyl)amido)(o-tolyl)(α-naphthalen-2-diyl(6-pyridin-2-diyl)methane)]hafnium dichloride,

[N-(2,6-di(1-methylethyl)phenyl)amido)(phenanthren-5-yl)(α-naphthalen-2-diyl(6-pyridin-2-diyl)methane)]hafnium dimethyl,

[N-(2,6-di(1-methylethyl)phenyl)amido)((henanthren-5-yl)(α-naphthalen-2-diyl(6-pyridin-2-diyl)methane)]hafnium di(N,N-dimethylamido), and

[N-(2,6-di(1-methylethyl)phenyl)amido)(phenanthren-5-yl)(α-naphthalen-2-diyl(6-pyridin-2-diyl)methane)]hafnium dichloride.

The foregoing metal complexes are conveniently prepared by standard metallation and ligand exchange procedures involving a source of the transition metal and the neutral ligand source. The last six listed complexes have been previously disclosed in WO 02/38628. Other techniques to prepare the foregoing complexes may be used as well.

Suitable activating cocatalysts useful in combination with component a) are those compounds capable of abstraction of a substituent therefrom to form an inert, noninterfering

counter ion, or that form a zwitterionic or other catalytically active derivative of a). Suitable activating cocatalysts for use herein include perfluorinated tri(aryl)boron compounds, and most especially tris(pentafluorophenyl)borane; nonpolymeric, compatible, noncoordinating, ion forming compounds (including the use of such compounds under oxidizing conditions), especially ammonium-, phosphonium-, oxonium-, carbonium-, silylium- or sulfonium- salts of compatible, noncoordinating anions, and ferrocenium salts of compatible, noncoordinating anions. A combination of the foregoing activating cocatalysts may be employed as well.

More particularly, suitable ion forming compounds useful as cocatalysts in one embodiment of the present invention comprise a cation which is a Bronsted acid capable of donating a proton, and a compatible, noncoordinating anion, A^-. Preferred anions are those containing a single coordination complex comprising a charge-bearing metal or metalloid core which anion is capable of balancing the charge of the active catalyst species (the metal cation) which may be formed when the two components are combined. Also, said anion should be sufficiently labile to be displaced by olefinic, diolefinic and acetylenically unsaturated compounds or other neutral Lewis bases such as ethers or nitriles. Suitable metals include, but are not limited to, aluminum, gold and platinum. Suitable metalloids include, but are not limited to, boron, phosphorus, and silicon. Compounds containing anions which comprise coordination complexes containing a single metal or metalloid atom are, of course, well known and many, particularly such compounds containing a single boron atom in the anion portion, are available commercially.

Preferably such cocatalysts may be represented by the following general formula:

$$(L^*\text{-}H)^+_d\,(A)^{d-}\quad (IX)$$

wherein:

L^* is a neutral Lewis base;

$(L^*\text{-}H)^+$ is a Bronsted acid;

A^{d-} is a noncoordinating, compatible anion having a charge of d-, and

d is an integer from 1 to 3.

More preferably A^{d-} corresponds to the formula: $[M'Q_4]^-$;

wherein:

M' is boron or aluminum in the +3 formal oxidation state; and

Q independently each occurrence is selected from hydride, dialkylamido, halide, hydrocarbyl, hydrocarbyloxide, halosubstituted-hydrocarbyl, hydroxy- substituted hydrocarbyl, halosubstituted hydrocarbyloxy, and halo- substituted silylhydrocarbyl radicals (including perhalogenated hydrocarbyl- perhalogenated hydrocarbyloxy- and perhalogenated silylhydrocarbyl radicals), said Q having up to 20 carbons with the proviso that in not more

than one occurrence is Q halide. Examples of suitable hydrocarbyloxide Q groups are disclosed in US-A-5,296,433.

In a more preferred embodiment, d is one, that is, the counter ion has a single negative charge and is A^-. Activating cocatalysts comprising boron which are particularly useful in the preparation of catalysts of this invention may be represented by the following general formula: $(L^*\text{-}H)^+(BQ_4)^-$;

wherein:

L* is as previously defined;

B is boron in a formal oxidation state of 3; and

Q is a hydrocarbyl-, hydrocarbyloxy-, fluorinated hydrocarbyl-, fluorinated hydrocarbyloxy-, or fluorinated silylhydrocarbyl- group of up to 20 nonhydrogen atoms, with the proviso that in not more than one occasion is Q hydrocarbyl.

Most preferably, Q is each occurrence a fluorinated aryl group, especially, a pentafluorophenyl group.

Illustrative, but not limiting, examples of boron compounds which may be used as an activating cocatalyst in the preparation of the improved catalysts of this invention are tri-substituted ammonium salts such as:

trimethylammonium tetrakis(pentafluorophenyl) borate,

triethylammonium tetrakis(pentafluorophenyl) borate,

tripropylammonium tetrakis(pentafluorophenyl) borate,

tri(n-butyl)ammonium tetrakis(pentafluorophenyl) borate,

tri(sec-butyl)ammonium tetrakis(pentafluorophenyl) borate,

N,N-dimethyl-N-dodecylammonium tetrakis(pentafluorophenyl) borate,

N,N-dimethyl-N-octadecylammonium tetrakis(pentafluorophenyl) borate,

N-methyl-N,N-didodecylammonium tetrakis(pentafluorophenyl) borate,

N-methyl-N,N-dioctadecylammonium tetrakis(pentafluorophenyl) borate,

N,N-dimethylanilinium tetrakis(pentafluorophenyl) borate,

N,N-dimethylanilinium n-butyltris(pentafluorophenyl) borate,

N,N-dimethylanilinium benzyltris(pentafluorophenyl) borate,

N,N-dimethylanilinium tetrakis(4-(t-butyldimethylsilyl)-2, 3, 5, 6-tetrafluorophenyl) borate,

N,N-dimethylanilinium tetrakis(4-(triisopropylsilyl)-2, 3, 5, 6-tetrafluorophenyl) borate,

N,N-dimethylanilinium pentafluorophenoxytris(pentafluorophenyl) borate,

N,N-diethylanilinium tetrakis(pentafluorophenyl) borate,

N,N-dimethyl-2,4,6-trimethylanilinium tetrakis(pentafluorophenyl) borate,

trimethylammonium tetrakis(2,3,4,6-tetrafluorophenyl)borate,

triethylammonium tetrakis(2,3,4,6-tetrafluorophenyl) borate,
tripropylammonium tetrakis(2,3,4,6-tetrafluorophenyl) borate,
tri(n-butyl)ammonium tetrakis(2,3,4,6-tetrafluorophenyl) borate,
dimethyl(t-butyl)ammonium tetrakis(2,3,4,6-tetrafluorophenyl) borate,
N,N-dimethylanilinium tetrakis(2,3,4,6-tetrafluorophenyl) borate,
N,N-diethylanilinium tetrakis(2,3,4,6-tetrafluorophenyl) borate, and
N,N-dimethyl-2,4,6-trimethylanilinium tetrakis(2,3,4,6-tetrafluorophenyl) borate;
disubstituted ammonium salts such as:
di-(i-propyl)ammonium tetrakis(pentafluorophenyl) borate, and
dicyclohexylammonium tetrakis(pentafluorophenyl) borate;
trisubstituted phosphonium salts such as:
triphenylphosphonium tetrakis(pentafluorophenyl) borate,
tri(o-tolyl)phosphonium tetrakis(pentafluorophenyl) borate, and
tri(2,6-dimethylphenyl)phosphonium tetrakis(pentafluorophenyl) borate;
disubstituted oxonium salts such as:
diphenyloxonium tetrakis(pentafluorophenyl) borate,
di(o-tolyl)oxonium tetrakis(pentafluorophenyl) borate, and
di(2,6-dimethylphenyl)oxonium tetrakis(pentafluorophenyl) borate;
disubstituted sulfonium salts such as:
diphenylsulfonium tetrakis(pentafluorophenyl) borate,
di(o-tolyl)sulfonium tetrakis(pentafluorophenyl) borate, and
bis(2,6-dimethylphenyl)sulfonium tetrakis(pentafluorophenyl) borate.

Preferred $(L^*\text{-}H)^+$ cations are N,N-dimethylanilinium, tributylammonium, N-methyl-N,N-di(dodecyl)ammonium, N-methyl-N,N-di(tetradecyl)ammonium, N-methyl-N,N-di(hexadecyl)ammonium, N-methyl-N,N-di(octadecyl)ammonium, and mixtures thereof. The latter three cations are the primary ammonium cations derived from a commercially available mixture of C_{14-18} tallow amines, and are collectively referred to as bis-hydrogenated tallowalkyl methylammonium cation. The resulting ammonium salt of the tetrakis(pentafluorophenyl)borate anion accordingly is know as bis-hydrogenated tallowalkyl methylammonium tetrakis(pentafluorophenyl)borate.

Another suitable ion forming, activating cocatalyst comprises a salt of a cationic oxidizing agent and a noncoordinating, compatible anion represented by the formula: $(Ox^{e+})_d(A^{d-})_e$.

wherein:

Ox^{e+} is a cationic oxidizing agent having a charge of e+;

e is an integer from 1 to 3; and

A^{d-} and d are as previously defined.

Examples of cationic oxidizing agents include: ferrocenium, hydrocarbyl-substituted ferrocenium, Ag^{+} or Pb^{+2}. Preferred embodiments of A^{d-} are those anions previously defined with respect to the Bronsted acid containing activating cocatalysts, especially tetrakis(pentafluorophenyl)borate.

Another suitable ion forming, activating cocatalyst comprises a compound which is a salt of a carbenium ion and a noncoordinating, compatible anion represented by the formula:

$\copyright^{+}A^{-}$

wherein:

$\copyright^{+}$ is a C_{1-20} carbenium ion; and

A^{-} is as previously defined. A preferred carbenium ion is the trityl cation, i.e. triphenylmethylium.

A further suitable ion forming, activating cocatalyst comprises a compound which is a salt of a silylium ion and a noncoordinating, compatible anion represented by the formula:

$R^{*}_{3}Si^{+}A^{-}$

wherein:

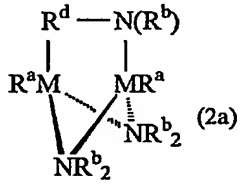
R^{*} is C_{1-10} hydrocarbyl, and A^{-} are as previously defined.

Preferred silylium salt activating cocatalysts are trimethylsilylium tetrakispentafluorophenylborate, triethylsilylium tetrakispentafluorophenylborate and ether substituted adducts thereof.

Certain complexes of alcohols, mercaptans, silanols, and oximes with tris(pentafluorophenyl)borane are also effective catalyst activators and may be used according to the present invention. Such cocatalysts are disclosed in USP 5,296,433, the teachings of which are herein incorporated by reference.

The most preferred activating cocatalysts are trispentafluorophenylborane and a mixture of long chain ammonium salts of tetrakis(pentafluorophenyl)borate, especially N,N-dioctadecyl-N-methylammonium tetrakpentafluorophenylborate, N-methyl-N,N-di(hexadecyl)ammonium tetrakpentafluorophenylborate and N,N-ditetradecyl-N-methylammonium tetrakpentafluorophenylborate. The latter mixture of borate salts is derived from hydrogenated tallow amine, and is referred to as bis-hydrogenated tallowalkyl methylammonium tetrakis(pentafluorophenyl)borate.

The Group 13 component, component c) of the catalyst composition of the invention, preferably corresponds to the formula $R^1Al(NR^2_2)_2$ wherein R^1 is C_{1-4} alkyl, and R^2 independently each occurrence is C_{6-20} aryl, preferably phenyl or a compound corresponding to the formula:



(2a)

wherein R^a is C_{1-4} alkyl, R^b is C_{6-20} aryl, preferably phenyl, and R^d is C_{6-20} arylene, preferably phenylene. A most highly preferred Group 13 component is bis(ethylaluminum)-1-phenylene-2-(phenyl)amido μ-bisdiphenylamide.

The process may be used to polymerize ethylenically unsaturated monomers having from 2 to 20 carbon atoms either alone or in combination. Preferred monomers include monovinylidene aromatic monomers, 4-vinylcyclohexene, vinylcyclohexane, norbornadiene and C_{2-10} aliphatic α-olefins (especially ethylene, propylene, isobutylene, 1-butene, 1-hexene, 3-methyl-1-pentene, 4-methyl-1-pentene, and 1-octene), C_{4-40} dienes, and mixtures thereof. Of the dienes typically used to prepare EPDMs, the particularly preferred dienes are 1,4-hexadiene (HD), 5-ethylidene-2-norbornene (ENB), 5-vinylidene-2-norbornene (VNB), 5-methylene-2-norbornene (MNB), and dicyclopentadiene (DCPD). The especially preferred dienes are 5-ethylidene-2-norbornene (ENB) and 1,4-hexadiene (HD). Most preferred monomers are ethylene, mixtures of ethylene, propylene and ethylidenenorbornene, or mixtures of ethylene and a C_{4-8} α-olefin, especially 1-octene.

In general, the polymerization may be accomplished at conditions well known in the prior art for Ziegler-Natta or Kaminsky-Sinn type polymerization reactions, that is, temperatures from 0-250 °C, preferably 30 to 200 °C and pressures from atmospheric to 30,000 atmospheres or higher. Suspension, solution, slurry, gas phase, solid state powder polymerization or other process condition may be employed if desired. A support, especially silica, alumina, or a polymer (especially poly(tetrafluoroethylene) or a polyolefin) may be employed, and desirably is employed when the catalysts are used in a gas phase polymerization process. The support is preferably employed in an amount to provide a weight ratio of catalyst (based on metal):support from 1:100,000 to 1:10, more preferably from 1:50,000 to 1:20, and most preferably from 1:10,000 to 1:30.

In most polymerization reactions the molar ratio of catalyst:polymerizable compounds employed is from 10^{-12}:1 to 10^{-1}:1, more preferably from 10^{-9}:1 to 10^{-5}:1.

Suitable solvents for polymerization are inert liquids. Examples include straight and branched-chain hydrocarbons such as isobutane, butane, pentane, hexane, heptane, octane, and mixtures thereof; cyclic and alicyclic hydrocarbons such as cyclohexane, cycloheptane, methylcyclohexane, methylcycloheptane, and mixtures thereof; perfluorinated hydrocarbons such as perfluorinated C_{4-10} alkanes, and the like and aromatic and alkyl-substituted aromatic compounds such as benzene, toluene, xylene, ethylbenzene and the like. Suitable solvents also include liquid olefins which may act as monomers or comonomers including ethylene, propylene, butadiene, cyclopentene, 1-hexene, 1-hexane, 4-vinylcyclohexene, vinylcyclohexane, 3-methyl-1-pentene, 4-methyl-1-pentene, 1,4-hexadiene, 1-octene, 1-decene, styrene, divinylbenzene, allylbenzene, vinyltoluene (including all isomers alone or in admixture), and the like. Mixtures of the foregoing are also suitable.

The catalysts may be utilized in combination with at least one additional homogeneous or heterogeneous polymerization catalyst in separate reactors connected in series or in parallel to prepare polymer blends having desirable properties.

Utilizing the catalyst compositions of the present invention copolymers having high comonomer incorporation and correspondingly low density, yet having a low melt index may be readily prepared. That is, high molecular weight polymers are readily attained by use of the present catalysts even at elevated reactor temperatures. This result is highly desirable because the molecular weight of α-olefin copolymers can be readily reduced by the use of hydrogen or similar chain transfer agent, however increasing the molecular weight of α-olefin copolymers is usually only attainable by reducing the polymerization temperature of the reactor. Disadvantageously, operation of a polymerization reactor at reduced temperatures significantly increases the cost of operation since heat must be removed from the reactor to maintain the reduced reaction temperature, while at the same time heat must be added to the reactor effluent to vaporize the solvent. In addition, productivity is increased due to improved polymer solubility, decreased solution viscosity, and a higher polymer concentration. Utilizing the present catalyst compositions, α-olefin homopolymers and copolymers having densities from 0.85 g/cm^3 to 0.96 g/cm^3, and melt flow rates from 0.001 to 10.0 dg/min are readily attained in a high temperature process.

The catalyst compositions of the present invention are particularly advantageous for the production of ethylene homopolymers and ethylene/α-olefin copolymers having high levels of long chain branching. The use of the catalyst compositions of the present invention

in continuous polymerization processes, especially continuous solution polymerization processes, allows for elevated reactor temperatures which favor the formation of vinyl terminated polymer chains that may be incorporated into a growing polymer, thereby giving a long chain branch. The use of the present catalyst compositions advantageously allows for the economical production of ethylene/α-olefin copolymers having processability similar to high pressure, free radical produced low density polyethylene.

As previously mentioned, the present catalyst composition is particularly useful in the preparation of EP and EPDM copolymers in high yield and productivity. The process employed may be either a solution or slurry process both of which are previously known in the art. Kaminsky, J. Poly. Sci., Vol. 23, pp. 2151-64 (1985) reported the use of a soluble bis(cyclopentadienyl) zirconium dimethyl-alumoxane catalyst system for solution polymerization of EP and EPDM elastomers. US-A-5,229,478 disclosed a slurry polymerization process utilizing similar bis(cyclopentadienyl) zirconium based catalyst systems.

The catalyst composition may be prepared as a homogeneous catalyst by addition of the requisite components to a solvent in which polymerization will be carried out by solution polymerization procedures. The catalyst composition may also be prepared and employed as a heterogeneous catalyst by adsorbing the requisite components on a catalyst support material such as silica gel, alumina or other suitable inorganic support material. When prepared in heterogeneous or supported form, it is preferred to use silica as the support material. The heterogeneous form of the catalyst system is employed in a slurry polymerization. As a practical limitation, slurry polymerization takes place in liquid diluents in which the polymer product is substantially insoluble. Preferably, the diluent for slurry polymerization is one or more hydrocarbons with less than 5 carbon atoms. If desired, saturated hydrocarbons such as ethane, propane or butane may be used in whole or part as the diluent. Likewise the α-olefin monomer or a mixture of different α-olefin monomers may be used in whole or part as the diluent. Most preferably the diluent comprises in at least major part the α-olefin monomer or monomers to be polymerized.

In contrast, solution polymerization conditions utilize a solvent for the respective components of the reaction, particularly the EP or EPDM polymer. Preferred solvents include mineral oils and the various hydrocarbons which are liquid at reaction temperatures. Illustrative examples of useful solvents include alkanes such as pentane, iso-pentane, hexane, heptane, octane and nonane, as well as mixtures of alkanes including kerosene and Isopar E™, available from Exxon Chemicals Inc.; cycloalkanes such as cyclopentane and

cyclohexane; and aromatics such as benzene, toluene, xylenes, ethylbenzene and diethylbenzene.

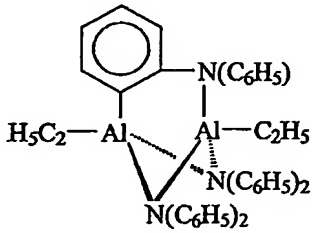
At all times, the individual ingredients as well as the recovered catalyst components must be protected from oxygen and moisture. Therefore, the catalyst components and catalysts must be prepared and recovered in an oxygen and moisture free atmosphere. Preferably, therefore, the reactions are performed in the presence of a dry, inert gas such as, for example, nitrogen.

Generally the polymerization process is carried out with a differential pressure of ethylene of from about 10 to about 1000 psi (70 to 7000 kPa), most preferably from about 40 to about 400 psi (30 to 300 kPa). The polymerization is generally conducted at a temperature of from 25 to 200 °C, preferably from 75 to 170 °C, and most preferably from greater than 95 to 160 °C.

The polymerization may be carried out as a batchwise or a continuous polymerization process. A continuous process is preferred, in which event the catalyst composition or the individual components thereof, monomer(s), and optionally solvent are continuously supplied to the reaction zone and polymer product continuously or semicontinuously removed therefrom.

The skilled artisan will appreciate that the invention disclosed herein may be practiced in the absence of any component which has not been specifically disclosed. The following examples are provided as further illustration of the invention and are not to be construed as limiting. Unless stated to the contrary all parts and percentages are expressed on a weight basis.

Example 1 Bis(ethylaluminum)-1-phenylene-2-(phenyl)amido μ-bisdiphenylamide



A 30 L glass reactor was charged with 12 L of toluene followed by 2.744 kg of a 25 percent solution of triethylaluminum in toluene. The solution was heated to 90 °C. In a dry box 2.034 kg of diphenylamine was dissolved in 8 L of toluene. This solution was slowly

added to the triethylaluminum solution over a period of three hours through a transfer line. After completion of the diphenylamine addition, the solution temperature was maintained between 95-105 °C for 72 hours. The resulting reaction mixture was cooled and transferred to a dry nitrogen purged receiver.

A portion of the resulting product was removed from the receiver, concentrated and a single crystal prepared for analysis by X-ray crystallography. The resulting structure (ORTEP) is shown in Figure 1.

Ethylene/1-Octene Copolymerization

A stirred 3.8 liter reactor was charged with about 1450 g of Isopar-E™ mixed alkanes solvent (available from Exxon Chemicals Inc.) and about 126 g of 1-octene comonomer. Hydrogen (10 mMol) was added as a molecular weight control agent using a mass flow meter. The reactor was heated to the polymerization temperature of 130 ºC and saturated with ethylene at 450 psig (3.1 MPa). Catalyst, (t-butylamido)dimethyl(η^5-tetramethylcyclopenta-dienyl)silanetitanium (II) η^4-1,3-pentadiene (A), and cocatalyst, trispentafluorophenylborane (FAB), were dissolved in Isopar E™ and premixed in a drybox with the aluminum tertiary component, and transferred to a catalyst addition system and injected into the reactor over approximately 3 minutes using a flow of high pressure Isopar E™ solvent. The polymerization conditions were maintained for 10 minutes with ethylene supplied on demand to maintain 450 psig reactor pressure. The ethylene consumed during the reaction was monitored using a mass flow meter and this consumption was used to calculate the catalyst efficiency.

High crystalline fraction (HCF) analysis was measured semi-quantitatively using the cooling curve of a differential scanning calorimetry scan for each polymer. The scan was examined to determine if the HCF peak appearing in the region from 75-78 °C was detectable and, if so, whether the peak was slight (detectable but not readily measurable) or significant (measurable). Then each 3rd component was given a HCF score based on the following criteria:

1. No detectable HCF
2. Slight to not detectable HCF
3. Significant to slight HCF
4. Significant HCF.

Results are contained in Table 1.

Table 1

Run	Catalyst (μmol)	Cocatalyst (μmol)	Aluminum compound (μmol)	C_2H_5 consumed (g)	Efficiency[1]	HCF[2]
1	2.50	7.50	25.0	71.3	28.52	1
2	2.50	7.50	0	15.9	6.36	4
3	4.50	13.50	0	44.8	9.96	4

1. grams of ethylene consumed per μmol of titanium.
2. high crystallinity fraction, semi-quantitive value

Ethylene/ Styrene Copolymerization

A stirred 3.8 liter reactor was charged with toluene and styrene comonomer. The reactor was sealed and heated to the polymerization temperature and saturated with ethylene at 275 psig (2.0 MPa). Catalyst, (t-butylamido)dimethyl(η^5-tetramethylcyclopenta-dienyl)silanetitanium (II) η^4-1,3-pentadiene (A), dimethyl[N-(1,1-dimethylethyl)-1,1-dimethyl-[1,2,3,4,5-η)-1,5,6,7-tetrahydro-2-methyl-s-indacen-1-yl]silanaminto(2-)-N] titanium (B), prepared according to the teachings of USP 5,965,756, or (1H-cyclopenta[*l*]phenanthrene-2-yl)dimethyl(t-butylamido)silanetitanium dimethyl, prepared according to the teachings of USP 6,150,297 (C), and cocatalyst, bis-hydrogenated tallowalkyl methylammonium tetrakis(pentafluorophenyl)borate (D) prepared according to the teachings of USP 5,919,983, or (trispentafluorophenylborane (E) or were dissolved in toluene and premixed in a drybox with the aluminum tertiary component, (bis(ethylaluminum)-1-phenylene-2-(phenyl)amido μ-bisdiphenylamide (F) or methylalumoxane (G)) and transferred to a catalyst addition system and injected into the reactor over approximately 1-2 minutes using a flow of high pressure solvent. The polymerization conditions were maintained for 10 minutes with ethylene supplied on demand to maintain reactor pressure. The polymer solution was discharged from the reactor into a nitrogen-purged glass kettle containing 200 mL of isopropanol and additive (IRGANOX 1010 and IRGAFOS 168). The polymer solution was poured into a tray, air dried overnight, then thoroughly dried in a vacuum oven for several days. The ethylene consumed during the reaction was monitored using a mass flow meter and used to calculate the catalyst efficiency. Results are contained in Table 2.

Table 2

Run	Cat. (μmol)	Cocat. (μmol)	Al comp. (μmol)	Temp. °C	C_2H_5 (MPa)	Toluene (g)	Styrene (g)	Eff.[1]	HCF
4	C (5)	D (5.5)	F (125)	110	2.2	1200	400	2.80	1
5	C (5)	D (5.5)	-	110	"	"	"	0.20	4
6	C (5)	E (7.5)	F(125)	110	"	"	"	20.64	1
7	C (5)	E (7.5)	-	110	"	"	"	5.14	4
8	B (18)	E (27)	F (180)	110	"	900	700	2.96	1
9	B (18)	E (27)	G (180)	110	"	"	"	0.08	3
10	A (10)	E (15)	F (100)	80	1.5	700	900	6.23	1
11	A (10)	E (15)	G (100)	80	1.5	"	"	0.13	3

[1] grams of ethylene consumed per μmol of titanium.

A: $[(\eta^5\text{-}Me_4C_5)SiMe_2N^tBu]Ti(\eta^4\text{-}1,3\text{-pentadiene})$ (CAS number 169104-71-6)

B: $[(\eta^5\text{-}MeC_{12}H_9)SiMe_2N^tBu]Ti(\eta^4\text{-}1,3\text{-pentadiene})$ (CAS numbers 199876-48-7 and 200074-30-2; mixture of isomers)

C: $[(\eta^5\text{-}C_{17}H_{10})SiMe_2N^tBu]TiMe_2$ (CAS number 221527-98-6)

D: $R_2N(H)Me\ B(C_6F_5)_4$, R = hydrogenated tallowalkyl (CAS number 200644-82-2)

E: $(C_6F_5)_3B$ (CAS number 1109-15-5)

F: Reaction product of Et_3Al with 2 molar equivalents of Ph_2NH according to Example 1

G: methylalumoxane (MMAO Type 3A, Akzo Nobel Company)

WHAT IS CLAIMED IS:

1. A catalyst composition comprising:

a) a transition metal complex capable of being activated for polymerization of addition polymerizable monomers;

b) an activator compound able to render the transition metal complex catalytically active for polymerization of addition polymerizable monomers; and

c) a Group 13 metal compound corresponding to the formula:

$$R^aM(N(R^b)_2)_2 \text{ (1) or } \begin{matrix} R^d\text{—}N(R^b) \\ | \quad\quad | \\ R^c_2M \quad MR^c_2 \end{matrix} \text{ (2);}$$

wherein,

M, independently each occurrence is a group 13 metal, preferably aluminum;

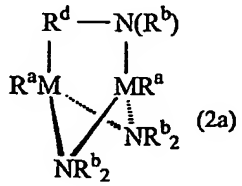
R^a is a hydrocarbyl, halocarbyl, halohydrocarbyl, tri(hydrocarbyl)silyl, or tri(hydrocarbyl)silyl- substituted hydrocarbyl radical of from 1 to 20 carbon, silicon or mixtures of carbon and silicon atoms, preferably C_{1-6} alkyl;

R^b independently each occurrence is a C_{1-30} hydrocarbyl group, preferably alkyl or aryl, most preferably C_{6-20} aryl;

R^c independently each occurrence is selected from hydrogen, R^a, $-NR^b_2$, or a halo- or di(C_{1-10} hydrocarbyl)amino- substituted hydrocarbyl group, and optionally one or more of R^c groups may be shared by both metal centers, M, in the form of a μ-bridged structure, preferably R^c is a hydrocarbyl group or $-NR^b_2$, wherein R^b is C_{6-20} aryl; and

R^d, is a divalent, anionic ligand group of up to 30 atoms, not counting hydrogen, preferably a hydrocarbadiyl group, or a halo- or di(C_{1-10} hydrocarbyl)amino- substituted hydrocarbadiyl group, most preferably R^d is C_{6-20} arylene.

2. A catalyst composition according to claim 1 wherein the Group 13 component corresponds to the formula $R^1Al(NR^2_2)_2$ wherein R^1 is C_{1-4} alkyl, and R^2 independently each occurrence is C_{6-20} aryl, preferably phenyl or a compound corresponding to the formula:



(2a)

wherein R^a is C_{1-4} alkyl, R^b is C_{6-20} aryl, preferably phenyl, and R^d is C_{6-20} arylene, preferably phenylene.

3. A catalyst composition according to claim 2 wherein the Group 13 component is bis(ethylaluminum)-1-phenylene-2-(phenyl)amido μ-bisdiphenylamide..

4. A catalyst composition according to claim 1 wherein the molar ratio of metal complex to component b) is from 1:1 to 1:50.

5. A catalyst composition according to claim 1 wherein the activating cocatalyst comprises trispentafluorophenylborane, N-methyl-N,N-dioctadecylammonium tetrakis(pentafluorophenyl)borate, or bis-hydrogenated tallowalkyl methylammonium tetrakis(pentafluorophenyl)borate.

6. A process for polymerization of addition polymerizable monomers or mixtures thereof comprising contacting said monomer or mixture of monomers with a catalyst system comprising the catalyst composition of claim 1 under addition polymerization conditions.

7. The process of claim 6 wherein the addition polymerizable monomer is a C_{2-20} α-olefin or a mixture thereof.

8. The process of claim 7 wherein ethylene and styrene are copolyerized.

ABSTRACT OF THE DISCLOSURE

Catalyst compositions that are highly tolerant of catalyst poisons for use in addition polymerizations comprising a catalytic derivative of a Group 4 metal complex, a cocatalyst, and a Group 13 metal amide compound.

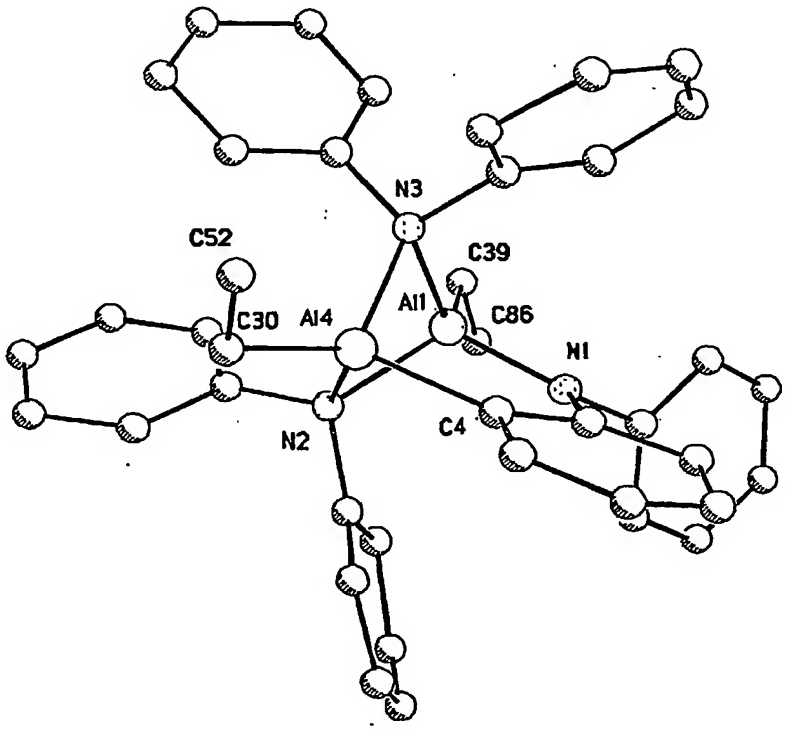


Figure 1

The Dow Chemical Company

Midland, Michigan 48674

May 3, 2005

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9-011-31-70-340-3016
THIS TRANSMISSION = 10 PAGES TOTAL

EUROPEAN PATENT OFFICE
P.B. 5818 - Patentlaan 2
NL-2280 HV Rijswijk
THE NETHERLANDS

RE: ENTRY INTO THE REGIONAL PHASE BEFORE THE EPO
FROM PCT APPLICATION NO. **PCT/US03/36483**
EPO APPLICATION NO. **03768952.8**
APPLICANT(S): **DOW GLOBAL TECHNOLOGIES INC.**
DEADLINE UNDER ARTICLE 39(1): **13 June 2005**
(Case No. **62619A**)

Dear Sir/Madam:

Enclosed are EPO Form 1200 (pages .1 through .5) to effect entry into the regional phase before the EPO for the above-identified International application. We have authorized Automatic Debit Order.

Form 1037 ("Acknowledgment of receipt for sender") is also enclosed, in triplicate.

We have named a registered European representation in Box No. 2 of EPO Form 1200.1. We have also enclosed an additional sheet naming additional representatives.

Very truly yours,

DOW GLOBAL TECHNOLOGIES INC.

By: Douglas N. Deline
Douglas N. Deline

cc: **BECK GREENER**

Fulwood House
12, Fulwood Place
London, England WC1V 6HR
UNITED KINGDOM

An das Europäische Patentamt
Nur für ab 1. Juli 1999 eingereichte internationale Anmeldungen!

To the European Patent Office
Only for international applications filed from 1 July 1999 onwards!

A l'office européen des brevets
Seulement pour les demandes internationales déposées à compter du 1er juillet 1999 !

1

Eintritt in die europäische Phase (EPA als Bestimmungsamt oder ausgewähltes Amt)

Entry into the European phase (EPO as designated or elected Office)

Entrée dans la phase européenne (l'OEB agissant en qualité d'Office désigné ou élu)

Europäische Anmeldenummer oder, falls nicht bekannt, PCT-Aktenzeichen oder PCT-Veröffentlichungsnummer

European application number, or, if not known, PCT application or publication number

03768952.8

Numéro de dépôt de la demande de brevet européen ou, à défaut, numéro de dépôt PCT ou de publication PCT

Zeichen des Anmelders oder Vertreters (max. 15 Positionen)

Applicant's or representative's reference (max. 15 spaces)

62619A

Référence du demandeur ou du mandataire (15 caractères ou espaces au maximum)

☒ **1. Anmelder**
Die Angaben über den (die) Anmelder sind in der internationalen Veröffentlichung enthalten oder vom Internationalen Büro nach der internationalen Veröffentlichung vermerkt worden.

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Indications concerning the applicant(s) are contained in the international publication or recorded by the International Bureau after the international publication.

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Les indications concernant le(s) demandeur(s) figurent dans la publication internationale ou ont été enregistrées par le Bureau international après la publication internationale.

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Changes which have not yet been recorded by the International Bureau are set out on an additional sheet.

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Zustellanschrift
(siehe Merkblatt II, 1)

Address for correspondence
(see *Notes II, 1)*

Adresse pour la correspondance
(voir notice II,1)

2. Vertreter

Name (Nur einen Vertreter angeben, der in das europäische Patentregister eingetragen und an den zugestellt wird)

2. Representative

Name (Name only one representative who will be listed in the Register of European Patents and to whom notification will be made)

John Raynor

2. Mandataire

Nom (N'indiquer qu' un seul mandataire, qui sera inscrit au Registre européen des brevets et auquel signification sera faite)

Geschäftsanschrift

Address of place of business

BECK GREENER , Fulwood House, 12, Fulwood Place, London, England, WC1V 6HR, United Kingdom

Adresse professionnelle

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Telephone

44-207-405-0921

Téléphone

Telefax **Telex**

Fax **Telex**

44-207-405-8113

Téléfax **Télex**

☒ Weitere(r) Vertreter auf Zusatzblatt

Additional representative(s) on additional sheet

Autre(s) mandataire(s) sur feuille additionnelle

3. Vollmacht

☐ Einzelvollmacht ist beigefügt.

3. Authorisation

Individual authorisation is attached.

3. Pouvoir

Un pouvoir spécial est joint.

☐ Allgemeine Vollmacht ist registriert unter Nummer:

General authorisation has been registered under No:

Un pouvoir général a été enregistré sous le n° :

☐ Allgemeine Vollmacht ist eingereicht, aber noch nicht registriert.

A general authorisation has been filed, but not yet registered.

Un pouvoir général a été déposé, mais n'est pas encore enregistré.

☐ Die beim EPA als PCT-Anmeldeamt eingereichte Vollmacht schließt ausdrücklich die europäische Phase ein.

The authorisation filed with the EPO as PCT receiving Office expressly includes the European phase.

Le pouvoir général déposé à l'OEB agissant en qualité d'office récepteur au titre du PCT s'applique expressément à la phase européenne.

[X]	**4. Prüfungsantrag** Hiermit wird die Prüfung der Anmeldung gemäß Art. 94 EPÜ beantragt. Die Prüfungsgebühr wird (wurde) entrichtet. *Prüfungsantrag in einer zugelassenen Nichtamtssprache (siehe Merkblatt III, 5.2):*	**4. Request for examination** Examination of the application under Art. 94 EPC is hereby requested. The examination fee is being (has been, will be) paid. *Request for examination in an admissible non-EPO language (see Notes III, 5.2):*	**4. Requête en examen** Il est demandé que soit examinée, la demande de brevet conformément à l'art. 94 CBE. Il est (a été, sera) procédé au paiement de la taxe d'examen. *Requête en examen dans une langue non officielle autorisée (voir notice III, 5.2):*
[ ]	**5. Abschriften** Zusätzliche Abschrift(en) der im ergänzenden europäischen Recherchenbericht angeführten Schriftstücke wird (werden) beantragt. Anzahl der zusätzlichen Sätze von Abschriften	**5. Copies** Additional copy (copies) of the documents cited in the supplementary European search report is (are) requested. Number of additional sets of copies	**5. Copies** Prière de fournir une ou plusieurs copies supplémentaires des documents cités dans le rapport complémentaire de recherche européenne. Nombre de jeux supplémentaires de copies
	6. Für das Verfahren vor dem EPA bestimmte Unterlagen	**6. Documents intended for proceedings before the EPO**	**6. Pièces destinées à la procédure devant l'OEB**
	6.1 Dem Verfahren vor dem EPA als Bestimmungsamt (PCT I) sind folgende Unterlagen zugrunde zu legen:	6.1 Proceedings before the EPO as designated Office (PCT I) are to be based on the following documents:	6.1 La procédure devant l'OEB agissant en qualité d'office désigné (PCT I) doit se fonder sur les pièces suivantes :
[ ]	die vom Internationalen Büro veröffentlichten Anmeldungsunterlagen (mit allen Ansprüchen, Beschreibung und Zeichnungen), gegebenenfalls mit den geänderten Ansprüchen nach Art. 19 PCT	the application documents published by the International Bureau (with all claims, description and drawings), where applicable with amended claims under Art. 19 PCT	les pièces de la demande publiée par le Bureau international (avec toutes les revendications, la description et les dessins), éventuellement avec les revendications modifiées conformément à l'article 19 du PCT
[ ]	soweit sie nicht ersetzt werden durch die beigefügten Änderungen.	unless replaced by the amendments enclosed.	dans la mesure où elles ne sont pas remplacées par les modifications jointes.
	Falls nötig, sind Klarstellungen auf einem Zusatzblatt einzureichen!	*Where necessary, clarifications must be submitted on a separate sheet!*	*Le cas échéant, des explications doivent être jointes sur une feuille additionnelle!*
	6.2 Dem Verfahren vor dem EPA als ausgewähltem Amt (PCT II) sind folgende Unterlagen zugrunde zu legen:	6.2 Proceedings before the EPO as elected Office (PCT II) are to be based on the following documents:	6.2 La procédure devant l'OEB agissant en qualité d'office élu (PCT II) doit se fonder sur les pièces suivantes :
[X]	die dem internationalen vorläufigen Prüfungsbericht zugrunde gelegten Unterlagen einschließlich seiner eventuellen Anlagen *(Solche Anlagen müssen immer beigefügt werden)*	the documents on which the international preliminary examination report is based, including its possible annexes *(Such annexes must always be filed)*	les pièces sur lesquelles se fonde le rapport d'examen préliminaire international, y compris ses annexes éventuelles *(De telles annexes sont toujours à joindre)*
[ ]	soweit sie nicht ersetzt werden durch die beigefügten Änderungen.	unless replaced by the amendments enclosed.	dans la mesure où elles ne sont pas remplacées par les modifications jointes.
	Falls nötig, sind Klarstellungen auf einem Zusatzblatt einzureichen!	*Where necessary, clarifications must be submitted on a separate sheet!*	*Le cas échéant, des explications doivent être jointes sur une feuille additionnelle!*
[X]	Sind dem EPA als mit der internationalen vorläufigen Prüfung beauftragten Behörde Versuchsberichte zugegangen, dürfen diese dem Verfahren vor dem EPA zugrunde gelegt werden.	If the EPO as International Preliminary Examining Authority has received test reports, these may be used as the basis of proceedings before the EPO.	Si l'OEB, agissant en qualité d'administration chargée de l'examen préliminaire international, a reçu des rapports d'essais, ceux-ci peuvent constituer la base de la procédure devant l'OEB.

	7. Übersetzungen Beigefügt sind die nachfolgend angekreuzten Übersetzungen in einer der Amtssprachen des EPA (Deutsch, Englisch, Französisch):	**7. Translations** Translations in one of the official languages of the EPO (English, French, German) are enclosed as crossed below:	**7. Traductions** Vous trouverez, ci-joint, les traductions cochées ci-après dans l'une des langues officielles de l'OEB (allemand, anglais, français):
	• *Im Verfahren vor dem EPA als Bestimmungsamt oder ausgewähltem Amt (PCT I+II):*	• *In proceedings before the EPO as designated or elected Office (PCT I + II):*	• *Dans la procédure devant l'OEB agissant en qualité d'Office désigné ou élu (PCT I + II):*
☐	Übersetzung der ursprünglich eingereichten internationalen Anmeldung (Beschreibung, Ansprüche, etwaige Textbestandteile in den Zeichnungen), der veröffentlichten Zusammenfassung, und etwaiger Angaben über biologisches Material nach Regel 13bis.3 und 13bis.4 PCT	Translation of the international application (description, claims, any text in the drawings) as originally filed, of the abstract as published and of any indication under Rule 13bis.3 and 13bis.4 PCT regarding biological material	Traduction de la demande internationale telle que déposée initialement (description, revendications, textes figurant éventuellement dans les dessins), de l'abrégé publié, et de toutes indications visées aux règles 13bis.3 et 13bis.4 du PCT concernant le matériel biologique
☐	Übersetzung der prioritätsbegründenden Anmeldung(en)	Translation of the priority application(s)	Traduction de la (des) demande(s) ouvrant le droit de priorité
☐	Es wird hiermit erklärt, daß die internationale Anmeldung in ihrer ursprünglich eingereichten Fassung eine vollständige Übersetzung der früheren Anmeldung ist (Regel 38(5) EPÜ)	It is hereby declared that the international application as originally filed is a complete translation of the previous application (Rule 38(5) EPC)	Il est déclaré par la présente que la demande internationale telle que déposée initialement est une traduction intégrale de la demande antérieure (règle 38(5) CBE)
	• *Zusätzlich im Verfahren vor dem EPA als Bestimmungsamt (PCT I):*	• *In addition, in proceedings before the EPO as designated Office (PCT I):*	• *De plus, dans la procédure devant l'OEB agissant en qualité d'office désigné (PCT I):*
☐	Übersetzung der nach Art. 19 PCT geänderten Ansprüche nebst Erklärung, falls diese dem Verfahren vor dem EPA zugrunde gelegt werden sollen (siehe Feld 6)	Translation of amended claims and any statement under Art. 19 PCT, if the claims as amended are to form the basis for the proceedings before the EPO (see Section 6)	Traduction des revendications modifiées et de la déclaration faite conformément à l'article 19 du PCT, si la procédure devant l'OEB doit être fondée sur les revendications modifiées (voir la rubrique 6)
	• *Zusätzlich im Verfahren vor dem EPA als ausgewähltem Amt (PCT II):*	• *In addition, in proceedings before the EPO as elected Office (PCT II):*	• *De plus, dans la procédure devant l'OEB agissant en qualité d'office élu (PCT II):*
☐	Übersetzung der Anlagen zum internationalen vorläufigen Prüfungsbericht	Translation of any annexes to the international preliminary examination report	Traduction des annexes du rapport d'examen préliminaire international
☐	**8. Biologisches Material** Die Erfindung bezieht sich auf bzw. Verwendet biologisches Material, das nach Regel 28 EPÜ hinterlegt worden ist.	**8. Biological material** The invention relates to and/or uses biological material deposited under Rule 28 EPC.	**8. Matière biologique** L'invention concerne et/ou utilise de la matière biologique, déposée conformément à la règle 28 CBE.
☐	Die Angaben nach Regel 28(1)c) EPÜ (falls noch nicht bekannt, die Hinterlegungsstelle und das (die) Bezugszeichen [Nummer, Symbole usw.] des Hinterlegers) sind in der internationalen Veröffentlichung oder in der gemäß Feld 7 eingereichten Übersetzung enthalten auf: Seite(n) / Zeile(n)	The particulars referred to in Rule 28(1)(c) EPC (if not yet known, the depository institution and the identification reference(s) [number, symbols etc.] of the depositor) are given in the international publication or in the translation submitted under Section 7 on: page(s) / line(s)	Les indications visées à la règle 28(1)c) CBE (si pas encore connues, l'autorité de dépôt et la (les) référence(s) d'identification [numéro ou symboles etc.] du déposant) figurent dans la publication internationale ou dans une traduction produite conformément à la rubrique 7 à la / aux page(s) / ligne(s)
	Die Empfangsbescheinigung(en) der Hinterlegungsstelle	The receipt(s) of deposit issued by the depositary institution	Le(s) récépissé(s) de dépôt délivré(s) par l'autorité de dépôt
☐	ist (sind) beigefügt	is (are) enclosed	est (sont) joint(s)
☐	wird (werden) nachgereicht	will be filed at a later date	sera (seront) produit(s) ultérieurement
☐	Verzicht auf die Verpflichtung des Antragstellers nach Regel 28(3) auf gesondertem Schriftstück	Waiver of the right to an undertaking from the requester pursuant to Rule 28(3) attached.	Renonciation, sur document distinct, à l'engagement du requérant au titre de la règle 28(3).

☐	**9.** **Nucleotid- und Aminosäure-sequenzen** Die nach Regeln 5.2 und 13[ter] PCT sowie Regel 111 (3) EPÜ erforderlichen Unterlagen liegen dem EPA bereits vor.	**9.** **Nucleotide and amino acid sequences** The items necessary in accordance with Rules 5.2 and 13[ter] PCT and Rule 111 (3) EPC have already been furnished to the EPO.	**9.** **Séquences de nucléotides et d'acides aminés** Les pièces requises selon les règles 5.2 et 13[ter] PCT et la règle 111 (3) CBE ont déjà été déposées auprès de l'OEB.
☐	Das schriftliche Sequenzprotokoll wird anliegend in einer Amtssprache des EPA nachgereicht.	The written sequence listing is furnished herewith in an official language of the EPO.	La liste de séquences écrite est produite ci-joint dans une des langues officielles de l'OEB.
☐	Das Sequenzprotokoll geht nicht über den Inhalt der Anmeldung in der ursprünglich eingereichten Fassung hinaus.	The sequence listing does not include matter which goes beyond the content of the application as filed.	La liste de séquences ne contient pas d'éléments s'étendant au-delà du contenu de la demande telle qu'elle a été déposée.
☐	Der vorgeschriebene Datenträger ist beigefügt.	The prescribed data carrier is enclosed.	Le support de données prescrit est joint.
☐	Die auf dem Datenträger gespeicherte Information stimmt mit dem schriftlichen Sequenzprotokoll überein.	The information recorded on the data carrier is identical to the written sequence listing.	L'information figurant sur le support de données est identique à celle que contient la liste de séquences écrite.
	10. **Benennungsgebühren***	**10.** **Designation fees***	**10.** **Taxes de désignation***
☒	10.1 Es ist derzeit beabsichtigt, den siebenfachen Betrag einer Benennungsgebühr zu entrichten. Damit gelten die Benennungsgebühren für alle Vertragsstaaten des EPÜ[1] als entrichtet (Art. 2 Nr. 3 GebO), soweit sie in der internationalen Anmeldung bestimmt sind.	10.1 It is currently intended to pay seven times the amount of the designation fee. The designation fees for all the EPC contracting states[1] designated in the international application are thereby deemed to have been paid (Art. 2 No. 3 Rfees).	10.1 Il est actuellement envisagé de payer un montant correspondant à sept fois la taxe de désignation. Les taxes de désignation sont ainsi réputées payées pour tous les Etats contractants de la CBE[1] désignés dans la demande internationale (art. 2, point 3 du RRT).
☐	10.2 Abweichend von der Erklärung in Nr. 10.1 ist derzeit beabsichtigt, weniger als sieben Benennungsgebühren für folgende in der internationalen Anmeldung bestimmte Vertragsstaaten des EPÜ[2] zu entrichten:	10.2 The declaration in No. 10.1 does not apply. Instead, it is currently intended to pay fewer than seven designation fees for the following EPC contracting states[2] designated in the international application:	10.2: Contrairement à ce qui est indiqué au n° 10.1, il est actuellement envisagé de payer moins de sept taxes de désignation pour les Etats contractants de la CBE[2] suivants désignés dans la demande internationale
	(1) ☐ _____ (4) ☐ _____ (2) ☐ _____ (5) ☐ _____ (3) ☐ _____ (6) ☐ _____		
	Soweit unter Nr. 10.2 Vertragsstaaten aufgeführt sind, wird beantragt, für die dort nicht aufgeführten Vertragsstaaten von der Zustellung einer Mitteilung nach Regel 108(3) EPÜ abzusehen.	If contracting states are indicated under No. 10.2, it is requested that no communication under Rule 108(3) EPC be issued for contracting states not thus indicated.	Si des Etats contractants sont mentionnés au n° 10.2, prière de ne pas procéder à la signification d'une notification prévue par les règle 108(3) CBE pour les Etats contractants n'ayant pas été y mentionnés.
	10.3 Wird ein automatischer Abbuchungsauftrag erteilt (Feld 12), so wird das EPA beauftragt, bei Ablauf der Grundfrist nach Regel 107(1)d) EPÜ den siebenfachen Betrag einer Benennungsgebühr abzubuchen. Ist eine Erklärung nach Nr. 10.2 abgegeben worden, so sollen die Benennungsgebühren nur für die dort angegebenen Vertragsstaaten abgebucht werden, sofern dem EPA nicht bis zum Ablauf der Grundfrist ein anderslautender Auftrag zugeht.	10.3 If an automatic debit order has been issued (Section 12), the EPO is authorised, on expiry of the basic period under Rule 107(1)(d) EPC, to debit seven times the amount of the designation fee. If states are indicated under No. 10.2, the EPO will debit designation fees only for those states, unless instructed otherwise before the basic period expires.	10.3 Si un ordre de prélèvement auto-matique est donné (rubrique 12), il est demandé à l'OEB de prélever, à l'expiration du délai normal visé à la règle 107(1)d) CBE, un montant correspondant à sept fois la taxe de désignation. Si une déclaration a été faite au n° 10.2, les taxes de désigna-tion ne sont à prélever que pour les Etats contractants qui y sont indiqués, sauf instruction contraire reçue par l'OEB avant l'expiration du délai normal.
	* Form 1200 (01.02) nur verwenden für internationale Anmeldungen, die ab 1. Juli 1999 eingereicht worden sind.	* Use Form 1200 (01.02) only for international applications filed from 1 July 1999 onwards.	* Veuillez utiliser le formulaire 1200 (01.02) seulement pour les demandes internationales déposées à compter du 1[er] juillet 1999.

1 Stand bei Drucklegung: 20 Vertragsstaaten, und zwar: / Status when this form was printed: 20 contracting states, namely / Situation à la date d'impression: 20 Etats contractants, à savoir: AT Österreich / Austria / Autriche, BE Belgien / Belgium / Belgique, CH/LI Schweiz und Liechtenstein / Switzerland and Liechtenstein / Suisse et Liechtenstein, CY Zypern / Cyprus / Chypre, DE Deutschland / Germany / Allemagne, DK Dänemark / Denmark / Danemark, ES Spanien / Spain / Espagne, FI Finnland / Finland / Finlande, FR Frankreich / France / France, GB Vereinigtes Königreich / United Kingdom / Royaume-Uni, GR Griechenland / Greece / Grèce, IE Irland / Ireland / Irlande, IT Italien / Italy / Italie, LU Luxemburg / Luxembourg / Luxembourg, MC Monaco / Monaco / Monaco, NL Niederlande / Netherlands / Pays-Bas, PT Portugal / Portugal / Portugal, SE Schweden / Sweden / Suède, TR Türkei / Turkey / Turquie

2 Für Türkei nur möglich, falls in der internationalen Anmeldung am oder nach dem 1. November 2000 bestimmt. / For Turkey possible only if designated in the international application on or after 1 November 2000. / En ce qui concerne Turquie, seulement si la désignation a été effectuée dans la demande internationale le 1[er] novembre 2000 ou à une date ultérieure.

☐	11.	Erstreckung des europäischen Patents Diese Anmeldung gilt auch als Erstreckungsantrag für alle in der internationalen Anmeldung bestimmten Nicht-Vertragsstaaten des EPÜ, mit denen bei Einreichung der internationalen Anmeldung »Erstrekkungsabkommen« in Kraft waren. Die Erstreckung wird jedoch nur wirksam, wenn die vorgeschriebene Erstreckungsgebühr entrichtet wird. Es ist derzeit beabsichtigt, die Erstreckungsgebühr für die nachfolgend angekreuzten Staaten zu entrichten:	11. Extension of the European patent This application is also considered as being a request for extension to all the non-Contracting States to the EPC designated in the international application with which "extension agreements" were in force on the date of filing the international application. However, the extension only takes effect if the prescribed extension fee is paid. It is currently intended to pay the extension fee for the States marked with a cross below:	11. Extension des effets du brevet européen La présente demande est également réputée demande d'extension à tous les Etats non contractants de la CBE désignés dans la demande internationale, avec lesquels existaient, lors du dépôt de la demande, des «accords d'extension». Toutefois l'extension ne produit ses effets que si la taxe d'extension prescrite est acquittée. Il est actuellement envisagé de payer la taxe d'extension pour les Etats dont le nom est coché ci-après:
☐	SI	Slowenien	Slovenia	Slovénie
☐	LT	Litauen	Lithuania	Lituanie
☐	LV	Lettland	Latvia	Lettonie
☐	AL	Albanien	Albania	Albanie
☐	RO	Rumänien	Romania	Roumanie
☐	MK	Ehemalige jugoslawische Republik Mazedonien	Former Yugoslav Republic of Macedonia	Ex-République yougoslave de Macédoine
☐		_____ 1)	_____ 1)	_____ 1)
	1)	Platz für Staaten, mit denen »Erstreckungsabkommen« nach Drucklegung dieses Formblatts in Kraft treten und die in der internationalen Anmeldung bestimmt waren.	1) Space for States with which "extension agreements" enter into force after this form has been printed and which were designated in the international application.	1) Prévu pour des Etats à l'égard desquels des « accords d'extension » entreront en vigueur après l'impression du présent formulaire et qui ont été désignés dans la demande internationale.
☒	12.	Automatischer Abbuchungsauftrag *(Nur möglich für Inhaber von beim EPA geführten laufenden Konten)* Das EPA wird beauftragt, nach Maßgabe der Vorschriften über das automatische Abbuchungsverfahren fällige Gebühren und Auslagen vom untenstehenden laufenden Konto abzubuchen. In Bezug auf die Benennungsgebühren wird auf Feld 10.3 verwiesen. Das EPA wird ferner beauftragt, die Erstreckungsgebühren für jeden in Feld 11 angekreuzten »Erstreckungsstaat« bis Ablauf der Grundfrist zu ihrer Zahlung abzubuchen, sofern ihm nicht bis dahin ein anderslautender Auftrag zugeht. Nummer und Kontoinhaber _____	12. Automatic debit order *(for EPO deposit account holders only)* The EPO is hereby authorised, under the Arrangements for the automatic debiting procedure, to debit from the deposit account below any fees and costs falling due. For designation fees, see Section 10.3. The EPO is also authorised, on expiry of the basic period for paying the extension fees, to debit those fees for each of the "extension states" marked with a cross in Section 11, unless instructed otherwise before the said period expires. Number and account holder 28300015 Dow	12. Ordre de prélèvement automatique *(uniquement possible pour les titulaires de comptes courants ouverts auprès de l'OEB)* Par la présente, il est demandé à l'OEB de prélever du compte courant ci-dessous les taxes et frais venant à échéance, conformément à la réglementation relative au prélèvement automatique. Pour les taxes de désignation, se reporter à la rubrique 10.3. Il est en outre demandé à l'OEB de prélever, à l'expiration du délai normal prévu pour leur paiement, les taxes d'extension pour chaque «Etat autorisant l'extension» coché à la rubrique 11, sauf instruction contraire reçue avant l'expiration de ce délai. Numéro et titulaire du compte _____
☒	13.	Eventuelle Rückzahlungen auf das beim EPA geführte laufende Konto Nummer und Kontoinhaber _____	13. Any reimbursement to EPO deposit account Number and account holder 28300015 Dow	13. Remboursements éventuels à effectuer sur le compte courant ouvert auprès de l'OEB Numéro et titulaire du compte _____
	14.	Unterschrift(en) des (der) Anmelder(s) oder Vertreters Ort / Datum	14. Signature(s) of applicant(s) or representative Place / Date	14. Signature(s) du (des) demandeur(s) ou du mandataire Lieu / Date

Signed at Midland, Michigan 48674, USA, **on** 27 April 2005

DOW GLOBAL TECHNOLOGIES INC.

By: Noreen D. Warrick

Noreen D. Warrick

Secretary

Authorized to act on behalf of Applicant

Für Angestellte (Art. 133(3) EPÜ) mit allgemeiner Vollmacht: Nr. _____	For employees (Art. 133(3) EPC) having a general authorisation No. _____	Pour les employés (art. 133(3) CBE) disposant d'un pouvoir général: N° _____
Name(n) des (der) Unterzeichneten bitte in Druckschrift wiederholen. Bei juristischen Personen bitte auch die Stellung des (der) Unterzeichneten innerhalb der Gesellschaft in Druckschrift angeben.	Please type name(s) under signature(s). In the case of legal persons, the position of the signatory within the company should also be printed.	Le ou les noms des signataires doivent être indiqués en caractères d'imprimerie. S'il s'agit d'une personne morale, la position occupée au sein de celle-ci par le ou les signataires doit également être indiquée en caractères d'imprimerie.

European Application Number **03768952.8**

PCT Application Number **PCT/US03/36483**

Applicant's reference **62619A**

Continuation of Box 2

Additional representatives are:

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1 Cover Letter to the EPO (Application No. 03768952.8; Dow Case No. 62619A);		
2 EPO Form 1200 to enter the regional phase before the EPO;		
3 Additional Sheet attached to EPO Form 1200		
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** Der Eingang der angegebenen Unterlagen wird bestätigt. Enthält diese Spalte keine Eintragungen, so wird lediglich bestätigt, daß eine Sendung zu dem angegebenen Aktenzeichen eingegangen ist.

* if already known

** The receipt of the items indicated is confirmed. If this column does not contain any entries, it is only confirmed that an item has been received for the indicated file.

* si déjà connu

** La réception des pièces indiquées est confirmée. Faute de mention dans cette colonne, le présent accusé de réception se rapporte à une pièce quelconque envoyée sous la référence indiquée.

EPA/EPO/OEB Form 1037.1 03.99
Eingangsbestätigung für Einsender
Acknowledgement of receipt for sender
Accusé de réception expéditeur

An das Europäische Patentamt	To the European Patent Office	A l'office européen des brevets
Nur für ab 1. Juli 1999 eingereichte internationale Anmeldungen!	*Only for international applications filed from 1 July 1999 onwards!*	*Seulement pour les demandes internationales déposées à compter du 1er juillet 1999 !*
Eintritt in die europäische Phase (EPA als Bestimmungsamt oder ausgewähltes Amt)	**Entry into the European phase (EPO as designated or elected Office)**	**Entrée dans la phase européenne (l'OEB agissant en qualité d'Office désigné ou élu)**

Europäische Anmeldenummer oder, falls nicht bekannt, PCT-Aktenzeichen oder PCT-Veröffentlichungsnummer	European application number, or, if not known, PCT application or publication number	Numéro de dépôt de la demande de brevet européen ou, à défaut, numéro de dépôt PCT ou de publication PCT
	03768952.8	
Zeichen des Anmelders oder Vertreters (max. 15 Positionen)	Applicant's or representative's reference (max. 15 spaces)	Référence du demandeur ou du mandataire (15 caractères ou espaces au maximum)
	62619A	

☒ **1. Anmelder**
Die Angaben über den (die) Anmelder sind in der internationalen Veröffentlichung enthalten oder vom Internationalen Büro nach der internationalen Veröffentlichung vermerkt werden.

☐ Änderungen, die das Internationale Büro noch nicht vermerkt hat, sind auf einem Zusatzblatt angegeben.

Zustellanschrift
(siehe Merkblatt II, 1)

1. Applicant
Indications concerning the applicant(s) are contained in the international publication or recorded by the International Bureau after the international publication.

Changes which have not yet been recorded by the International Bureau are set out on an additional sheet.

Address for correspondence
(see *Notes II, 1)*

1. Demandeur
Les indications concernant le(s) demandeur(s) figurent dans la publication internationale ou ont été enregistrées par le Bureau international après la publication internationale.

Les changements qui n'ont pas encore été enregistrés par le Bureau international sont indiqués sur une feuille additionelle.

Adresse pour la correspondance
(voir notice II,1)

2. Vertreter

Name (Nur einen Vertreter angeben, der in das europäische Patentregister eingetragen und an den zugestellt wird)

EPO - DG 1

Geschäftsanschrift

06. 05. 2005

Telefon

Telefax Telex

(109)

☒ Weitere(r) Vertreter auf Zusatzblatt

2. Representative

Name (Name only one representative who will be listed in the Register of European Patents and to whom notification will be made)

John Raynor

Address of place of business

BECK GREENER , Fulwood House, 12, Fulwood Place, London, England, WC1V 6HR, United Kingdom

Telephone

44-207-405-0921

Fax Telex

44-207-405-8113

Additional representative(s) on additional sheet

2. Mandataire

Nom (N'indiquer qu' un seul mandataire, qui sera inscrit au Registre européen des brevets et auquel signification sera faite)

Adresse professionnelle

Téléphone

Téléfax Télex

Autre(s) mandataire(s) sur feuille additionnelle

3. Vollmacht

☐ Einzelvollmacht ist beigefügt.

☐ Allgemeine Vollmacht ist registriert unter Nummer:

☐ Allgemeine Vollmacht ist eingereicht, aber noch nicht registriert.

☐ Die beim EPA als PCT-Anmeldeamt eingereichte Vollmacht schließt ausdrücklich die europäische Phase ein.

3. Authorisation

Individual authorisation is attached.

General authorisation has been registered under No:

A general authorisation has been filed, but not yet registered.

The authorisation filed with the EPO as PCT receiving Office expressly includes the European phase.

3. Pouvoir

Un pouvoir spécial est joint.

Un pouvoir général a été enregistré sous le n° :

Un pouvoir général a été déposé, mais n'est pas encore enregistré.

Le pouvoir général déposé à l'OEB agissant en qualité d'office récepteur au titre du PCT s'applique expressément à la phase européenne.

[X]	**4. Prüfungsantrag** Hiermit wird die Prüfung der Anmeldung gemäß Art. 94 EPÜ beantragt. Die Prüfungsgebühr wird (wurde) entrichtet. *Prüfungsantrag in einer zugelassenen Nichtamtssprache (siehe Merkblatt III, 5.2):*	**4. Request for examination** Examination of the application under Art. 94 EPC is hereby requested. The examination fee is being (has been, will be) paid. *Request for examination in an admissible non-EPO language (see Notes III, 5.2):*	**4. Requête en examen** Il est demandé que soit examinée, la demande de brevet conformément à l'art. 94 CBE. Il est (a été, sera) procédé au paiement de la taxe d'examen. *Requête en examen dans une langue non officielle autorisée (voir notice III, 5.2):*
☐	**5. Abschriften** Zusätzliche Abschrift(en) der im ergänzenden europäischen Recherchenbericht angeführten Schriftstücke wird (werden) beantragt. Anzahl der zusätzlichen Sätze von Abschriften	**5. Copies** Additional copy (copies) of the documents cited in the supplementary European search report is (are) requested. Number of additional sets of copies	**5. Copies** Prière de fournir une ou plusieurs copies supplémentaires des documents cités dans le rapport complémentaire de recherche européenne. Nombre de jeux supplémentaires de copies
	6. Für das Verfahren vor dem EPA bestimmte Unterlagen	**6. Documents intended for proceedings before the EPO**	**6. Pièces destinées à la procédure devant l'OEB**
	6.1 Dem Verfahren vor dem EPA als Bestimmungsamt (PCT I) sind folgende Unterlagen zugrunde zu legen:	6.1 Proceedings before the EPO as designated Office (PCT I) are to be based on the following documents:	6.1 La procédure devant l'OEB agissant en qualité d'office désigné (PCT I) doit se fonder sur les pièces suivantes :
☐	die vom Internationalen Büro veröffentlichten Anmeldungsunterlagen (mit allen Ansprüchen, Beschreibung und Zeichnungen), gegebenenfalls mit den geänderten Ansprüchen nach Art. 19 PCT	the application documents published by the International Bureau (with all claims, description and drawings), where applicable with amended claims under Art. 19 PCT	les pièces de la demande publiée par le Bureau international (avec toutes les revendications, la description et les dessins), éventuellement avec les revendications modifiées conformément à l'article 19 du PCT
☐	soweit sie nicht ersetzt werden durch die beigefügten Änderungen.	unless replaced by the amendments enclosed.	dans la mesure où elles ne sont pas remplacées par les modifications jointes.
	Falls nötig, sind Klarstellungen auf einem Zusatzblatt einzureichen !	*Where necessary, clarifications must be submitted on a separate sheet!*	*Le cas échéant, des explications doivent être jointes sur une feuille additionnelle!*
	6.2 Dem Verfahren vor dem EPA als ausgewähltem Amt (PCT II) sind folgende Unterlagen zugrunde zu legen:	6.2 Proceedings before the EPO as elected Office (PCT II) are to be based on the following documents:	6.2 La procédure devant l'OEB agissant en qualité d'office élu (PCT II) doit se fonder sur les pièces suivantes :
[X]	die dem internationalen vorläufigen Prüfungsbericht zugrunde gelegten Unterlagen einschließlich seiner eventuellen Anlagen *(Solche Anlagen müssen immer beigefügt werden)*	the documents on which the international preliminary examination report is based, including its possible annexes *(Such annexes must always be filed)*	les pièces sur lesquelles se fonde le rapport d'examen préliminaire international, y compris ses annexes éventuelles *(De telles annexes sont toujours à joindre)*
☐	soweit sie nicht ersetzt werden durch die beigefügten Änderungen.	unless replaced by the amendments enclosed.	dans la mesure où elles ne sont pas remplacées par les modifications jointes.
	Falls nötig, sind Klarstellungen auf einem Zusatzblatt einzureichen!	*Where necessary, clarifications must be submitted on a separate sheet!*	*Le cas échéant, des explications doivent être jointes sur une feuille additionnelle!*
[X]	Sind dem EPA als mit der internationalen vorläufigen Prüfung beauftragten Behörde Versuchsberichte zugegangen, dürfen diese dem Verfahren vor dem EPA zugrunde gelegt werden.	If the EPO as International Preliminary Examining Authority has received test reports, these may be used as the basis of proceedings before the EPO.	Si l'OEB, agissant en qualité d'administration chargée de l'examen préliminaire international, a reçu des rapports d'essais, ceux-ci peuvent constituer la base de la procédure devant l'OEB.

	7. Übersetzungen Beigefügt sind die nachfolgend angekreuzten Übersetzungen in einer der Amtssprachen des EPA (Deutsch, Englisch, Französisch):	**7. Translations** Translations in one of the official languages of the EPO (English, French, German) are enclosed as crossed below:	**7. Traductions** Vous trouverez, ci-joint, les traductions cochées ci-après dans l'une des langues officielles de l'OEB (allemand, anglais, français):
	• *Im Verfahren vor dem EPA als **Bestimmungsamt oder ausgewähltem Amt** (PCT I+II):*	• *In proceedings before the EPO **as designated or elected Office** (PCT I + II):*	• *Dans la procédure devant l'OEB **agissant en qualité d'Office désigné ou élu** (PCT I + II):*
☐	Übersetzung der **ursprünglich eingereichten internationalen Anmeldung** (Beschreibung, Ansprüche, etwaige Textbestandteile in den Zeichnungen), der veröffentlichten Zusammenfassung, und etwaiger Angaben über biologisches Material nach Regel 13bis.3 und 13bis.4 PCT	Translation of the **international application** (description, claims, any text in the drawings) as **originally filed**, of the abstract as published and of any indication under Rule 13bis.3 and 13bis.4 PCT regarding biological material	Traduction de la **demande internationale telle que déposée initialement** (description, revendications, textes figurant éventuellement dans les dessins), de l'abrégé publié, et de toutes indications visées aux règles 13bis.3 et 13bis.4 du PCT concernant le matériel biologique
☐	Übersetzung der **prioritätsbegründenden Anmeldung(en)**	Translation of the **priority application(s)**	Traduction de la (des) **demande(s) ouvrant le droit de priorité**
☐	Es wird hiermit erklärt, daß die internationale Anmeldung in ihrer ursprünglich eingereichten Fassung eine vollständige Übersetzung der früheren Anmeldung ist (Regel 38(5) EPÜ)	It is hereby declared that the international application as originally filed is a complete translation of the previous application (Rule 38(5) EPC)	Il est déclaré par la présente que la demande internationale telle que déposée initialement est une traduction intégrale de la demande antérieure (règle 38(5) CBE)
	• ***Zusätzlich** im Verfahren vor dem EPA als **Bestimmungsamt** (PCT I):*	• ***In addition**, in proceedings before the EPO as **designated Office** (PCT I):*	• ***De plus**, dans la procédure devant l'OEB agissant en qualité d'**office désigné** (PCT I):*
☐	Übersetzung der nach Art. 19 PCT **geänderten Ansprüche** nebst Erklärung, falls diese dem Verfahren vor dem EPA zugrunde gelegt werden sollen (siehe Feld 6)	Translation of **amended claims** and any statement under Art. 19 PCT, if the claims as amended are to form the basis for the proceedings before the EPO (see Section 6)	Traduction des **revendications modifiées** et de la déclaration faite conformément à l'article 19 du PCT, si la procédure devant l'OEB doit être fondée sur les revendications modifiées (voir la rubrique 6)
	• ***Zusätzlich** im Verfahren vor dem EPA als **ausgewähltem Amt** (PCT II):*	• ***In addition**, in proceedings before the EPO as **elected Office** (PCT II):*	• ***De plus**, dans la procédure devant l'OEB agissant en qualité d'**office élu** (PCT II):*
☐	Übersetzung der **Anlagen zum internationalen vorläufigen Prüfungsbericht**	Translation of any annexes to the **international preliminary examination report**	Traduction des **annexes du rapport d'examen préliminaire international**
☐	**8. Biologisches Material** Die Erfindung bezieht sich auf bzw. Verwendet biologisches Material, das nach Regel 28 EPÜ hinterlegt worden ist.	**8. Biological material** The invention relates to and/or uses biological material deposited under Rule 28 EPC.	**8. Matière biologique** L'invention concerne et/ou utilise de la matière biologique, déposée conformément à la règle 28 CBE.
☐	Die **Angaben nach Regel 28(1)c) EPÜ** (falls noch nicht bekannt, die Hinterlegungsstelle und das (die) Bezugszeichen [Nummer, Symbole usw.] des Hinterlegers) sind in der internationalen Veröffentlichung oder in der gemäß Feld 7 eingereichten Übersetzung enthalten auf:	The particulars **referred to in Rule 28(1)(c) EPC** (if not yet known, the depository institution and the identification reference(s) [number, symbols etc.] of the depositor) are given in the international publication or in the translation submitted under Section 7 on:	Les **indications visées à la règle 28(1)c) CBE** (si pas encore connues, l'autorité de dépôt et la (les) référence(s) d'identification [numéro ou symboles etc.] du déposant) figurent dans la publication internationale ou dans une traduction produite conformément à la rubrique 7 à la / aux
	Seite(n) / Zeile(n)	page(s) / line(s)	page(s) / ligne(s)
	Die **Empfangsbescheinigung(en)** der Hinterlegungsstelle	The **receipt(s) of deposit** issued by the depositary institution	**Le(s) récépissé(s) de dépôt** délivré(s) par l'autorité de dépôt
☐	ist (sind) beigefügt	is (are) enclosed	est (sont) joint(s)
☐	wird (werden) nachgereicht	will be filed at a later date	sera (seront) produit(s) ultérieurement
☐	Verzicht auf die Verpflichtung des Antragstellers nach Regel 28(3) auf gesondertem Schriftstück	Waiver of the right to an undertaking from the requester pursuant to Rule 28(3) attached.	Renonciation, sur document distinct, à l'engagement du requérant au titre de la règle 28(3).

☐	**9. Nucleotid- und Aminosäuresequenzen** Die nach Regeln 5.2 und 13ter PCT sowie Regel 111 (3) EPÜ erforderlichen Unterlagen liegen dem EPA bereits vor.	**9. Nucleotide and amino acid sequences** The items necessary in accordance with Rules 5.2 and 13ter PCT and Rule 111 (3) EPC have already been furnished to the EPO.	**9. Séquences de nucléotides et d'acides aminés** Les pièces requises selon les règles 5.2 et 13ter PCT et la règle 111 (3) CBE ont déjà été déposées auprès de l'OEB.
☐	Das schriftliche Sequenzprotokoll wird anliegend in einer Amtssprache des EPA nachgereicht.	The written sequence listing is furnished herewith in an official language of the EPO.	La liste de séquences écrite est produite ci-joint dans une des langues officielles de l'OEB.
☐	Das Sequenzprotokoll geht nicht über den Inhalt der Anmeldung in der ursprünglich eingereichten Fassung hinaus.	The sequence listing does not include matter which goes beyond the content of the application as filed.	La liste de séquences ne contient pas d'éléments s'étendant au-delà du contenu de la demande telle qu'elle a été déposée.
☐	Der vorgeschriebene Datenträger ist beigefügt.	The prescribed data carrier is enclosed.	Le support de données prescrit est joint.
☐	Die auf dem Datenträger gespeicherte Information stimmt mit dem schriftlichen Sequenzprotokoll überein.	The information recorded on the data carrier is identical to the written sequence listing.	L'information figurant sur le support de données est identique à celle que contient la liste de séquences écrite.

	10. Benennungsgebühren*	**10. Designation fees***	**10. Taxes de désignation***
☒	10.1 Es ist derzeit beabsichtigt, den **siebenfachen** Betrag einer Benennungsgebühr zu entrichten. Damit gelten die Benennungsgebühren für **alle Vertragsstaaten des EPÜ**[1] als entrichtet (Art. 2 Nr. 3 GebO), soweit sie **in der internationalen Anmeldung** bestimmt sind.	10.1 It is currently intended to pay **seven times** the amount of the designation fee. The designation fees for **all the EPC contracting states**[1] **designated in the international application** are thereby deemed to have been paid (Art. 2 No. 3 Rfees).	10.1 Il est actuellement envisagé de payer un montant correspondant à **sept fois** la taxe de désignation. Les taxes de désignation sont ainsi réputées payées pour **tous les Etats contractants de la CBE**[1] **désignés dans la demande internationale** (art. 2, point 3 du RRT).
☐	10.2 Abweichend von der Erklärung in Nr. 10.1 ist derzeit beabsichtigt, **weniger als sieben** Benennungsgebühren für folgende **in der internationalen Anmeldung bestimmte Vertragsstaaten des EPÜ**[2] zu entrichten:	10.2 The declaration in No. 10.1 does not apply. Instead, it is currently intended to pay **fewer than seven** designation fees for the **following EPC contracting states**[2] **designated in the international application:**	10.2: Contrairement à ce qui est indiqué au n° 10.1, il est actuellement envisagé de payer **moins de sept** taxes de désignation pour les **Etats contractants de la CBE**[2] **suivants désignés dans la demande internationale**
	(1) ☐ _____ (2) ☐ _____ (3) ☐ _____		(4) ☐ _____ (5) ☐ _____ (6) ☐ _____
	Soweit unter Nr. 10.2 Vertragsstaaten aufgeführt sind, wird beantragt, für die dort nicht aufgeführten Vertragsstaaten von der Zustellung einer Mitteilung nach Regel 108(3) EPÜ abzusehen.	If contracting states are indicated under No. 10.2, it is requested that no communication under Rule 108(3) EPC be issued for contracting states not thus indicated.	Si des Etats contractants sont mentionnés au n° 10.2, prière de ne pas procéder à la signification d'une notification prévue par les règle 108(3) CBE pour les Etats contractants n'ayant pas été y mentionnés.
	10.3 Wird ein **automatischer Abbuchungsauftrag** erteilt (Feld 12), so wird das EPA beauftragt, bei Ablauf der Grundfrist nach Regel 107(1)d) EPÜ den siebenfachen Betrag einer Benennungsgebühr abzubuchen. Ist eine Erklärung nach Nr. 10.2 abgegeben worden, so sollen die Benennungsgebühren nur für die dort angegebenen Vertragsstaaten abgebucht werden, sofern dem EPA nicht bis zum Ablauf der Grundfrist ein anderslautender Auftrag zugeht.	10.3 If an **automatic debit order** has been issued (Section 12), the EPO is authorised, on expiry of the basic period under Rule 107(1)(d) EPC, to debit seven times the amount of the designation fee. If states are indicated under No. 10.2, the EPO will debit designation fees only for those states, unless instructed otherwise before the basic period expires.	10.3 Si un ordre de prélèvement auto-matique est donné (rubrique 12), il est demandé à l'OEB de prélever, à l'expiration du délai normal visé à la règle 107(1)d) CBE, un montant correspondant à sept fois la taxe de désignation. Si une déclaration a été faite au n° 10.2, les taxes de désigna-tion ne sont à prélever que pour les Etats contractants qui y sont indiqués, sauf instruction contraire reçue par l'OEB avant l'expiration du délai normal.

* Form 1200 (01.02) nur verwenden für internationale Anmeldungen, die ab 1. Juli 1999 eingereicht worden sind.

* Use Form 1200 (01.02) only for international applications filed from 1 July 1999 onwards.

* Veuillez utiliser le formulaire 1200 (01.02) seulement pour les demandes internationales deposees a compter du 1er juillet 1999.

1 Stand bei Drucklegung: 20 Vertragsstaaten, und zwar: / Status when this form was printed: 20 contracting states, namely / Situation à la date d'impression: 20 Etats contractants, à savoir: AT Österreich / Austria / Autriche, BE Belgien / Belgium / Belgique, CH/LI Schweiz und Liechtenstein / Switzerland and Liechtenstein / Suisse et Liechtenstein, CY Zypern / Cyprus / Chypre, DE Deutschland / Germany / Allemagne, DK Dänemark / Denmark / Danemark, ES Spanien / Spain / Espagne, FI Finnland / Finland / Finlande, FR Frankreich / France / France, GB Vereinigtes Königreich / United Kingdom / Royaume-Uni, GR Griechenland / Greece / Grèce, IE Irland / Ireland / Irlande, IT Italien / Italy / Italie, LU Luxemburg / Luxembourg / Luxembourg, MC Monaco / Monaco / Monaco, NL Niederlande / Netherlands / Pays-Bas, PT Portugal / Portugal / Portugal, SE Schweden / Sweden / Suède, TR Türkei / Turkey / Turquie

2 Für Türkei nur möglich, falls in der internationalen Anmeldung am oder nach dem 1. November 2000 bestimmt. / For Turkey possible only if designated in the international application on or after 1 November 2000. / En ce qui concerne Turquie, seulement si la désignation a été effectuée dans la demande internationale le 1er novembre 2000 ou à une date ultérieure.

☐	**11. Erstreckung des europäischen Patents** Diese Anmeldung gilt auch als Erstreckungsantrag für alle in der internationalen Anmeldung bestimmten **Nicht-Vertragsstaaten** des EPÜ, mit denen bei Einreichung der internationalen Anmeldung »Erstrekkungsabkommen« in Kraft waren. Die Erstreckung wird jedoch nur wirksam, wenn die vorgeschriebene Erstreckungsgebühr entrichtet wird. Es ist derzeit beabsichtigt, die Erstreckungsgebühr für die nachfolgend angekreuzten Staaten zu entrichten:	**11. Extension of the European patent** This application is also considered as being a request for extension to all the **non-Contracting States** to the EPC designated in the international application with which "extension agreements" were in force on the date of filing the international application. However, the extension only takes effect if the prescribed extension fee is paid. It is currently intended to pay the extension fee for the States marked with a cross below:	**11. Extension des effets du brevet européen** La présente demande est également réputée demande d'extension à tous les **Etats non contractants** de la CBE désignés dans la demande internationale, avec lesquels existaient, lors du dépôt de la demande, des «accords d'extension». Toutefois l'extension ne produit ses effets que si la taxe d'extension prescrite est acquittée. Il est actuellement envisagé de payer la taxe d'extension pour les Etats dont le nom est coché ci-après:
☐	SI Slowenien	Slovenia	Slovénie
☐	LT Litauen	Lithuania	Lituanie
☐	LV Lettland	Latvia	Lettonie
☐	AL Albanien	Albania	Albanie
☐	RO Rumänien	Romania	Roumanie
☐	MK Ehemalige jugoslawische Republik Mazedonien	Former Yugoslav Republic of Macedonia	Ex-République yougoslave de Macédoine
☐	_____ 1)	_____ 1)	_____ 1)
	1) Platz für Staaten, mit denen »Erstreckungsabkommen« nach Drucklegung dieses Formblatts in Kraft treten und die in der internationalen Anmeldung bestimmt waren.	1) Space for States with which "extension agreements" enter into force after this form has been printed and which were designated in the international application.	1) Prévu pour des Etats à l'égard desquels des « accords d'extension» entreront en vigueur après l'impression du présent formulaire et qui ont été désignés dans la demande internationale.
☒	**12. Automatischer Abbuchungsauftrag** ***(Nur möglich für Inhaber von beim EPA geführten laufenden Konten)*** Das EPA wird beauftragt, nach Maßgabe der Vorschriften über das automatische Abbuchungsverfahren fällige Gebühren und Auslagen vom untenstehenden laufenden Konto abzubuchen. In Bezug auf die **Benennungsgebühren** wird auf Feld 10.3 verwiesen. Das EPA wird ferner beauftragt, die **Erstreckungsgebühren** für jeden in Feld 11 angekreuzten »Erstreckungsstaat« bis Ablauf der Grundfrist zu ihrer Zahlung abzubuchen, sofern ihm nicht bis dahin ein anderslautender Auftrag zugeht. Nummer und Kontoinhaber _____	**12. Automatic debit order** ***(for EPO deposit account holders only)*** The EPO is hereby authorised, under the Arrangements for the automatic debiting procedure, to debit from the deposit account below any fees and costs falling due. For **designation fees**, see Section 10.3. The EPO is also authorised, on expiry of the basic period for paying the extension fees, to debit those fees for each of the "extension states" marked with a cross in Section 11, unless instructed otherwise before the said period expires. Number and account holder **28300015 Dow**	**12. Ordre de prélèvement automatique** ***(uniquement possible pour les titulaires de comptes courants ouverts auprès de l'OEB)*** Par la présente, il est demandé à l'OEB de prélever du compte courant ci-dessous les taxes et frais venant à échéance, conformément à la réglementation relative au prélèvement automatique. Pour les **taxes de désignation**, se reporter à la rubrique 10.3. Il est en outre demandé à l'OEB de prélever, à l'expiration du délai normal prévu pour leur paiement, les **taxes d'extension** pour chaque «Etat autorisant l'extension» coché à la rubrique 11, sauf instruction contraire reçue avant l'expiration de ce délai. Numéro et titulaire du compte _____
☒	**13. Eventuelle Rückzahlungen** auf das beim EPA geführte laufende Konto Nummer und Kontoinhaber _____	**13. Any reimbursement to EPO deposit account** Number and account holder **28300015 Dow**	**13. Remboursements éventuels à effectuer sur le compte courant ouvert auprès de l'OEB** Numéro et titulaire du compte _____
	14. Unterschrift(en) des (der) Anmelder(s) oder Vertreters Ort / Datum	**14. Signature(s) of applicant(s) or representative Place / Date**	**14. Signature(s) du (des) demandeur(s) ou du mandataire Lieu / Date**

Signed at Midland, Michigan 48674, USA, **on** 27 April 2005

DOW GLOBAL TECHNOLOGIES INC.

By: Noreen D. Warrick

Noreen D. Warrick

Secretary

Authorized to act on behalf of Applicant

Für Angestellte (Art. 133(3) EPÜ) mit allgemeiner Vollmacht: **Nr.** _____	**For employees (Art. 133(3) EPC) having a general authorisation** **No.** _____	**Pour les employés (art. 133(3) CBE) disposant d'un pouvoir général:** **N°** _____
Name(n) des (der) Unterzeichneten bitte in Druckschrift wiederholen. Bei juristischen Personen bitte auch die Stellung des (der) Unterzeichneten innerhalb der Gesellschaft in Druckschrift angeben.	Please type name(s) under signature(s). In the case of legal persons, the position of the signatory within the company should also be printed.	Le ou les noms des signataires doivent être indiqués en caractères d'imprimerie. S'il s'agit d'une personne morale, la position occupée au sein de celle-ci par le ou les signataires doit également être indiquée en caractères d'imprimerie.

European Application Number **03768952.8**

PCT Application Number **PCT/US03/36483**

Applicant's reference **62619A**

Continuation of Box 2

Additional representatives are:

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(109)

DOW

The Dow Chemical Company

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May 3, 2005

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EUROPEAN PATENT OFFICE
P.B. 5818 - Patentlaan 2
NL-2280 HV Rijswijk
THE NETHERLANDS

RE: ENTRY INTO THE REGIONAL PHASE BEFORE THE EPO
FROM PCT APPLICATION NO. **PCT/US03/36483**
EPO APPLICATION NO. **03768952.8**
APPLICANT(S): **DOW GLOBAL TECHNOLOGIES INC.**
DEADLINE UNDER ARTICLE **39(1): 13 June 2005**
(Case No. **62619A**)

Dear Sir/Madam:

Enclosed are EPO Form 1200 (pages .1 through .5) to effect entry into the regional phase before the EPO for the above-identified International application. We have authorized Automatic Debit Order.

Form 1037 ("Acknowledgment of receipt for sender") is also enclosed, in triplicate.

We have named a registered European representation in Box No. 2 of EPO Form 1200.1. We have also enclosed an additional sheet naming additional representatives.

Very truly yours,

DOW GLOBAL TECHNOLOGIES INC.

By: Douglas N. Deline
Douglas N. Deline

EPO - DG 1
06. 05. 2005
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cc: **BECK GREENER**

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